

July 10, 2003

Popular Summary of Book Chapter for
Intercontinental Transport - Andreas Stohl, Editor
Springer-Verlag, Publisher

CHAPTER 7 by A. M. Thompson

TITLE: Intercontinental Transport of Ozone from Tropical Biomass Burning

Researchers have been looking at the connection between tropical biomass burning and ozone formation and long-range transport for roughly 15 years. One can see the linkage and transport patterns from satellite though aircraft and/or balloon-sonde profiles are required to observe the fine structure (ozone transport over thousands of km often happens in thin layers).

In this review, I survey the pyrogenic ozone transport in the large oceanic basins - Indian Ocean, Pacific and Atlantic. Mechanistic complexities are discussed and examples shown from satellite, aircraft and soundings, including NASA results from TOMS, the GTE experiments and the SHADOZ sounding program. Experiments referred to include SAFARI-92, TRACE-A, INDOEX, PEM-Tropics and TRACE-P.

Over the Atlantic it has become apparent that ozone pollution from biomass fires is augmented by subsidence, a variable tropopause height, and lightning - even ozone pollution from the Indian Ocean has been implicated. Over the Indian Ocean, pollution interacts with convection and the monsoon cycle.

Intercontinental Transport of Ozone from Tropical Biomass Burning

7.1 Introduction - Historical Perspective

The atmospheric impacts of tropical fires came to attention in the 1970's, largely through the work of Crutzen, in relationship to anthropogenic carbon dioxide and other long-lived greenhouse gases [Crutzen et al., 1979; Seiler and Crutzen, 1980]. It was realized that photochemically reactive gases released by fires (e.g. NO, CO, volatile organic carbon [VOC]) would interact as they do in an urban environment to form ozone, another greenhouse gas. Several sets of observations stimulated research into the relationship between biomass fires and ozone. A Brazilian field campaign in the 1980 dry season measured ozone and emissions of reactive gases [Crutzen et al., 1985; Delany et al., 1985]. Surface measurements made in west Africa were among the first to demonstrate a seasonality in ozone [Cros et al., 1988] that could be explained by savanna fires.

It was not right away that the long-range impact of biomass burning was apparent. The 1980 Brazilian campaign (and another in 1985) and early African ozone measurements were performed near source regions. Aircraft measurements of ozone during TROPOZ and STRATOZ, taken over the Atlantic, pointed to transport of ozone and of precursors like CO and other biomass burning byproducts, eg carboxylic acids in tropical precipitation (xxrefs). Along the Brazilian coast, the ozone sounding record at Natal (6S, 35W) displayed a seasonal enhancement in free tropospheric ozone consistent with transport from fire activity in southern Africa [Logan and Kirchhoff, 1986]. How could this happen? Through a combination of convective mixing of ozone and/or its precursors followed by advection in relatively stable air parcels [Chatfield and Delany, 1990; Pickering et al., 1992; Chatfield et al., 1996 Figure 7.1-1).

Although some of the early 1980's observations were suggestive, it was remote sensing and aircraft experiments remote from sources (e.g. FOS/DECAFE; TROPOZ II, SAFARI/TRACE-A; PEM-Tropics A) that revealed the far-ranging impact of biomass burning on ozone and its precursor carbon monoxide (CO). Carbon monoxide measured from space (an instrument aboard the Space Shuttle) in October 1984 and again in October 1994 showed widespread enhancements in this gas over large oceanic regions in the tropics [Reichle et al., 1986; Connors et al., 1999]. More recently, a three-year record of continuous CO measurements from the MOPITT instrument aboard NASA's Terra Spacecraft have confirmed locations and seasonality of the Shuttle snapshots of pyrogenic CO between 55 N/S [need MOPITT ref]. Chatfield et al. [2002] points out that commercial airliner (GASP = Global Atmospheric Sampling Project) CO measurements would have alerted us to tropical impacts of biomass burning in the 1970's had the quality of the GASP data been more certain.

The big push to connect ozone and long-range transport came from Fishman and coworkers [1990; 1991], who estimated the tropical tropospheric ozone (TTO) column amount by taking the difference between total ozone and stratospheric column ozone measured by two satellites. The resulting climatological maps showed a large seasonal maximum over the south tropical Atlantic occurring in austral Spring when southern hemisphere fire activity is very high. A plume also exits Africa toward the Indian Ocean in September-October-November (Figure 7.1-2). A large multi-national, multi-platform (6-8 aircraft and numerous ground-based experiments in half a dozen countries) called SAFARI-92 (Southern African Fire Atmospheric Research

Initiative) was assembled to study ecological aspects of fires, trace gas emissions from savanna fires, and near and far impacts of fire activity on atmospheric composition [*Journal of Geophysical Research*, **101**, 30 October 1996 issue; Andreae et al., IGAC book; van Wilgen et al., 1997]. Compendia of studies related to biomass burning and the atmosphere prior to and following SAFARI-92 and TRACE-A are given in Levine (1991) and (1995), respectively.

A partner experiment to SAFARI-92, TRACE-A (Transport and Atmospheric Chemistry near the Equator - Atlantic), tested the hypothesis that biomass burning is responsible for the south Atlantic ozone maximum. SAFARI/TRACE-A, like TROPOZ and FOS/DECAFE(?), confirmed the photochemical origins of much of the Atlantic ozone. Table 7-1 (Chap 5?) shows the "smog"-type reactions involved in ozone formation. Underlined are those species measured during various experiments (Table 7-2) that contributed to an understanding of ozone photochemistry and biomass burning sources. Other important measurements were made during aircraft sampling flights during the 1997 El-Nino-induced Indonesian fires [Matsueda et al., 1999].

In other recent observations, both in-situ and space-borne, a more complex picture of chemical and dynamical interactions affecting tropical ozone is emerging. By complexity is meant (a) regional meteorological patterns and variability and (b) on the global scale, interaction with large-scale meteorological patterns and processes that complicate what might otherwise be straightforward intercontinental flows. (Krishnamurti et al. [1996] deserve credit for pointing this out). These complexities are captured to varying degrees in models, as described in Chapters by Schultz and Bey (Chap 5) and by Lawrence (Chap 6). In the following Sections, one should bear in mind the conclusions of the model studies, as well as tropical observations reported about South Asia (Lawrence/ Chapter 6), Akimoto et al. (Chapter 4) and Piketh (Chapter 9). We attempt here to synthesize observations about the intercontinental transport of ozone from biomass fires by moving through successive large impact regions: Indian Ocean (Section 7.2), Pacific (Section 7.3), Atlantic (Section 7.4).

7.2 Indian Ocean Region

Pollution over the tropical Indian Ocean is affected principally by four source regions: Africa, south central Asia (mostly India), southeast Asia, the maritime continent (major components - Indonesia, New Guinea, Australia). In the following sections the relationship between each of these regions to trans-oceanic and intercontinental transport over the Indian Ocean is illustrated.

7.2.1 African Pollution

Examples of export transport pathways from southern Africa to the Indian Ocean appear in Chatfield et al [2002] and Piketh et al., [2002]. In Chatfield et al. [2002], the pattern indicated by a simulated CO plume coincides with elevated smoke aerosol observed in September 1996 (Figure 7.2-1) by the TOMS satellite. From various indicators (Southern Oscillation Index, Outgoing Longwave Radiation, Indian Dipole Mode index, precipitation and wind fields), as well as from satellite tropospheric ozone and aerosol measurements [Thompson et al., 2001], it appears that 1996 was a typical year. Ozone pollution traveled to the western Pacific where it was observed from aircraft during PEM-Tropics A (see Fenn et al., 1999; Gregory et al., 1999; Hoell et al., 1999; Stoller et al., 1999). Satellite fire count data [Olson et al., 1999] from the AVHRR (Advanced Very High Resolution Radiometer) instrument for the PEM-Tropics A period (28 August to 6 October 1996) shows that the highest distribution of fires was in the region shaded in Figure 7.2-1.

What did ozone over the Indian Ocean look like in Sept. 1996? Are the patterns suggested by the CO simulation of Chatfield et al. [2002] applicable to ozone observations? We address this question with ozone profile data from the SHADOZ (Southern Hemisphere Additional Ozonesondes; Thompson et al., 2003) stations at Réunion Island and Watukosek.

Figure 7.2-2 shows ozone profiles from soundings taken over La Réunion on 29 August and 9 October 1996. Radiosondes launched at the same time (not shown) reveal a capped marine boundary layer below 800 hPa. No pollution appears in the marine boundary layer (ozone mixing ratio is 20-30 ppbv, Figure 7.2-2). Air-parcel back trajectories (800 hPa, red, Figure 7.2-3a) are located over the ocean south of Africa. Between 400 and 700 hPa ozone mixing ratios from 50 to 115 ppbv suggest pollution (Figure 7.2-2). This is confirmed by air parcel back trajectories (blue, purple in Figure 7.2-3) that originate over regions of active fires (compare fire locations in Figure 7.2-1).

Fig. 7.2-4 shows profiles over Watukosek (Java) in July and September 1996. Pollution in the free troposphere (ozone > 50 ppbv at 300-600 hPa) appears to originate from eastern Africa and the western Indian Ocean (Figure 7.2-5). Note that on 17 September 1996 there is also relatively high ozone near the surface; this might be due to localized biomass fires and/or transport from northern Australian fires (trajectories at 700, 800 hPa in Figure 7.2-5b; fires shown in Figure 7.2-1).

How typical are these results? Ozone sonde observations at these stations, based on the SHADOZ record (Figure 5 in Thompson et al., 2003) confirm that this is the time of year of greatest tropospheric column ozone. For Réunion, column-integrated tropospheric ozone amounts appear in Figure 9 in Baldy et al. [1996] and Figure 2 in Taupin et al. [1999]. ✓
Fujiwara/2000 Watukosek Climo

The reason for highest ozone over Réunion in October is transport of ozone and/or ozone precursors from African and Madagascar fires [Baldy et al., 1996], although there are also stratospheric influences [Taupin et al., 1999; Randriambelo et al., 1999]. Trajectories [Baldy et al., 1996] and wind climatologies over Réunion [Taupin et al., 1999] show that above 4 km (~700 hPa) during September-October flows are predominantly from the southwest. Transit times from Madagascar and southeastern Africa are typically 1-2 and 4-5 days, respectively. A cross-section of mean ozone (mixing ratio as function of month and altitude from five years of SHADOZ data, Figure 7.2-6a) shows elevated concentrations in September-October-November where flows are usually African (3-10 km). Some of this ozone could come from nitric oxide injected by lightning because the ITCZ is approaching Réunion at this time of year. At Watukosek, in addition to ozone exported from Africa, there may be influences of biomass burning in Southeast Asia, the Indonesian maritime continent (see Sec. 7.2.3) and Australia. Figure 7.26b shows relatively high surface ozone at Watukosek in September-October [Fujiwara et al., 2000].

7.2.2 South and Southeast Asian Pollution

Intercontinental transport relative to South Asian and southeast Asian sources was explored during INDOEX (Indian Ocean Experiment), a dedicated campaign that considered many processes and worked downwind of India. Working during January-March 1999, during the period of winter monsoon-to-spring transition, INDOEX was a multi-platform, multi-national experiment. The reader is referred to a collection of INDOEX papers in the *Journal of Geophysical Research* (date) and to overview papers by Lelieveld et al. (2001) and Ramanathan et al. (2001). Highlights of intercontinental transport from south and SE Asia during INDOEX

are covered by Lawrence in Chapter 6. This section presents examples of satellite and station data that put the campaign data in perspective.

Figures 3.1 and 4.5 in Chapter 6 show flows governing pollution transport from south Asia toward the central Indian Ocean region. The large-scale circulation (arrows in Figure 7.2-7) which interacts with the pollutants includes downslope flow of ozone from the Himalayas. Although ozone pollution episodes during INDOEX were frequent, discriminating between pyrogenic and industrial sources requires analysis with tracers and modeling. In Phadnis et al [2002], evaluation of typical budgets is made with a coupled chemistry-transport model driven with winds from a GCM. In the free troposphere (represented by 685 hPa, Figure 10 in Phadnis et al., 2002), maximum ozone occurs over northern India. The fraction of pollution ozone, however, is greatest over SE India and the tropical Indian Ocean. This is due to a combination of subsidence with low-level outflow away from the continent. Similarly, the region of highest fractional pollution ozone in the lower-free troposphere due to SE Asian pollution extends over Indonesia and the Indian Ocean (Figure 11 in Phadnis et al., 2002). Based on the model, approximately half the ozone pollution is from biomass/biofuels burning and half from fossil fuel combustion.

The INDOEX experiment was timed to take advantage of maximum impact of south Asian emissions on the Indian Ocean. In ozone, this is reflected as layers of ozone > 40 ppbv in the free troposphere. Figure 7.2-8 presents contrasting ozone profiles from Male (Maldives Islands), an Indian Ocean site that intermittently receives south Asian pollution. The two soundings taken during INDOEX (cf Figure 11b in Thompson et al., 2003) correspond to contrasting flows near the southern Indian pollution source region (Figure 7.2-9). The ozone-enriched layers at 750 hPa and 500 hPa on 31 January 1999 appear to represent dehydrated continental air, traceable to south India and a localized region of pollution recirculation, respectively. In contrast, on 3 February, the lower free troposphere is moist with ozone mixing ratios of 20 ppbv; the flows are marine.

7.2.3 Indonesian Maritime and Australian Continents

deLaat and Lelieveld (2002) showed that SE Asian pollution tends to be transported more strongly toward the western Pacific than to the Indian Ocean. However, biomass burning in Indonesia and Australia, which often follows the El-Nino-La-Nina cycle, may affect pollution transport over the Indian Ocean (Komala et al., 1996; Fujiwara et al., 1999; Kita et al., 2000). The impact of biomass burning on ozone formation have been evaluated with satellite, aircraft and sounding data.

Figure 7.2-10 displays satellite tropospheric ozone and smoke aerosol for 10 October 1997 during the period of intense biomass fires over Kalimantan and New Guinea, the latter signified by gray shaded regions. The smoke is greatest near regions of burning. However, the ozone, most concentrated above the boundary layer [Thompson et al., 2001], clearly shows intercontinental transport. Ozone from Africa extends east to Australia and ozone from the Indonesian fires reaches southern India and beyond.

The BIBLE-A campaign (Kondo et al., JGR 107, 2002a) conducted in late Sept-early Oct 1998 was in a building La Nina when biomass burning pollution was relatively low over Indonesia (Kita et al., 2002). Sampling over the Indonesian maritime continent, New Guinea and Australia gave evidence of intercontinental transport from Indonesia toward the central Indian Ocean. Convection introduced ozone precursors (from non-biomass burning pollution and lightning) into the upper troposphere where ozone formed at a 1-2 ppbv/day rate. Convective

outflow toward the south introduced ozone into the westerlies (Figure 7.2-11) so the net flow was from Indonesia to northern Australia. The lightning input was greater in 1998 than during BIBLE-B in September-October 1999. Subsidence brought ozone over Australia and prevented northern Australian biomass burning from reaching the altitudes required for long-range transport.

7.3 Pacific Region

The Pacific is the largest oceanic basin affected by intercontinental-scale transport of ozone from biomass burning. The PEM (Pacific Exploratory Mission) series of experiments (1991, 1994, 1996, 1999) turned up evidence of pollution transport throughout the tropical and mid-latitude Pacific in all seasons [JGR Special Issue refs]. Figure 7.3-1 shows a climatology of tropospheric ozone over three Pacific stations that receive ozone from biomass burning sources. As Oltmans et al [2001] have pointed out, the highest concentrations of free tropospheric ozone (5-10 km) occur in September-October-November (SON) when outflow from southern hemisphere biomass burning is greatest. This can be seen in the contours for each station in Figure 7.3-1.

The greatest source of biomass burning emissions at Samoa and Fiji is from Africa, although ozone from SE Asian and Australian fires can also be detected. Individual ozone profiles with mean mid-tropospheric (5-10 km) ozone greater than 55 ppbv at these two stations appear in Figure 7.3-2a,b. All but four of the profiles depicted are from October and November. Two features are noteworthy. First, as Thompson et al. [2003] have noted, ozone is highly variable in the southern tropical troposphere. Many profiles display very clean (mixing ratio < 20 ppbv) layers as well as polluted layers even during the time of year that pollution is most prevalent. The persistent and widespread polluted ozone layers over the Pacific have been noted by Newell et al [1999] and Stoller et al [1999]. The PEM-Tropics A experiment conducted in September-October 1996 (*Journal of Geophysical Research*, **104**, March 20, 1999; *Journal of Geophysical Research*, **106**, July 20, 1999) measured a number of tracers (CO, hydrocarbons, PAN, formaldehyde, organic acids) to show the biomass burning origins of ozone.

Second, Fiji clearly receives more pollution influence than Samoa (Figures 8, 9 in Thompson et al., 2003). There is a clustering of high values (~70 ppbv) at 4-6 km over Fiji. This contributes to the generally higher ozone contour in SON at Fiji compared to Samoa (45 ppbv vs 35 ppbv in Figure 7.3-1). Back trajectories showing air parcel origins for the ozone profiles in Figure 7.3-2a,b are illustrated in Figure 7.3-3. Both Australian and African fires may be contributing to the ozone layers at Fiji and Samoa (cf fire locations in Figure 7.2-1).

At San Cristobal (Figure 7.3-1c) the July-October period is characterized by a broad ozone feature with mean mixing ratio contour of 45 ppbv. This appears at somewhat greater altitude (6-8 km; profiles in Figure 7.3-3a) than the corresponding features over Fiji and Samoa (Figures 7.3-1a,b). The trajectories that give origins of the highest ozone layers show the likely reason for the elevation (Figure 7.3-3b). The pollution originates from regions of South American burning. During the latter part of the dry season, ozone and ozone precursors near fires are entrained into developing convective systems, in either a cook-then-mix or mix-then-cook mode (Chatfield and Delany, 1990). These air parcels are then advected east toward the Pacific or into the westerlies toward the Atlantic or Africa [Thompson et al., 1996; Longo et al., 1999]. An example of high ozone over the Galapagos that is traced to high-ozone, biomass burning regions of South America is described by Oltmans et al., (2001; Plate 6).

Need to add relevant papers from TRACEP...

7.4 Atlantic Region

7.4.1 Intercontinental Transport from Africa and South America

As described in the Introduction, much of our knowledge about intercontinental transport of ozone pollution is based on observations over the tropical Atlantic and adjacent continents. Flows from biomass burning regions over Africa (Figure 7.1-2) and South America (Figure 7.3-4) can supply ozone in long-lived layers. The lifetime of ozone (and some ozone precursors) above the boundary layer is up to several weeks. For example, peroxyacetyl nitrate (PAN) is a reservoir for the essential ozone precursor NO [Schultz et al., 1999]. During transit from Africa, transformation of PAN to NO allows ozone to form far from source regions. Figure 7.1-2 shows the patterns of ozone (and ozone precursor) transport toward the south tropical Atlantic and the photochemical source from biomass fires schematically when the fires are south of the ITCZ. This applies to the July through October period. As the dry season advances south [Cahoon et al., 1992; Justice et al., 1996; Duncan et al., 2002/3??], so do the regions of most intense fire activity. During TRACE-A (September-October 1992), the highest ozone formation rates were found in the boundary layer near fires (10-15 ppbv/day ozone) and above 8 km where NO, presumably enriched by lightning [Smyth et al., 1996], maintained a positive photochemical regime [Thompson et al., 1996]. The upper tropospheric lightning influence on ozone occurred over the south tropical Atlantic and both adjacent continents.

In the middle troposphere (5-10 km) during TRACE-A (September-October 1992) high ozone layers (mean > 65 ppbv) were associated with recirculation about the south tropical Atlantic gyre. SAFARI-92 (also conducted during September-October 1992), SAFARI-2000 (August-September 2000) and related climatological analyses demonstrated how persistent are some "absolutely stable layers" of ozone pollution [Cosijn and Tyson, 1996; Garstang et al., 1996; Tyson et al., 1997; Freiman?? Thompson et al., 2002]. Recirculation of ozone over the persistent south tropical Atlantic high-pressure region allows ozone column amounts > 35 Dobson Units (one Dobson Unit, DU, is equivalent to 0.1 cm ozone thickness at one standard atmosphere) to remain intact for weeks at a time. Analysis with air parcel back trajectories showed that the high ozone over Ascension Island and Natal [Thompson et al., 1996] usually originated over Africa. Figure 7.4-1, with August and October ozone profiles at Natal and Ascension, respectively, from the SHADOZ record (2001 shown), is similar to the SAFARI-92/TRACE-A period [Thompson et al., 1996]. Likewise, analysis with air parcel back-trajectories illustrates African origins for these soundings (Figure 7.4-2).

Besides photochemical activity, several week lifetime and advection of ozone, there is a tendency for ozone to accumulate over the Atlantic due to subsidence associated with sinking motions of the Walker circulation [Krishnamurti et al., 1996; also Figure 18 in Thompson et al., 1996]. The result is sometimes a tropospheric ozone column more than 50 DU thickness. Atlantic subsidence was apparent in the ozone curtains from the uv-DIAL aircraft instrument during TRACE-A [Browell et al., 1996]. It also shows in the cross-section of mean monthly mixing ratio vs altitude (Figure 7.4-3) at the SHADOZ stations at Ascension, Natal and Paramaribo. For Ascension and Natal (Figure 7.4-3a,b) intercontinental transport is signified by the localized ozone maximum (55 ppbv contour in June-August; 85 ppbv contour in September-October) between 5 and 10 km. At Ascension a secondary maximum (65 ppbv) appears in February. At this time of year, burning north of the ITCZ in western Africa is prevalent [Figure 12 in Thompson, 2003] but some interhemispheric transport of ozone to Ascension, south of the ITCZ, is probably occurring (see Section 7.4.2).

North of the ITCZ, long-range ozone transport patterns are distinct from south of the

ITCZ. This shows in several ways. First, the analysis of the total ozone wave-one pattern by Kim et al. [1996] and Hudson and Thompson [1998] reveals that it is largely a southern hemisphere phenomenon. Second, consider the ozone vertical and seasonal distribution over Paramaribo [Figure 7.4-3c; also Peters et al., 2003]. In SON, there is relatively high ozone in the free troposphere over Paramaribo but not as distinct and pronounced as the 5-10 km high-ozone feature observed over Natal (10 degrees south of Paramaribo, and 20 degrees east). All the elevated ozone from June through March appears connected with the upper troposphere-lower stratosphere layer. During MAM relatively low ozone in the upper troposphere (a localized 35 ppbv minimum at 12 km) looks as if convective redistribution of boundary layer ozone to the upper troposphere occurs as the ITCZ passes over Surinam. Where do the high-ozone mid-tropospheric ozone layers over Paramaribo come from? Figure 7.4-4, from Peters et al. [2003], shows wind fields at 500 hPa and regions affected by biomass fires in early August 2000; as a consequence the ozone profile from 9 August 2000 was polluted.

Looking at Figures 7.4-3 (Atlantic), 7.2-3, 7.2-4 (Watukosek) and Figure 7.3-1 (Samoa) show why there is a zonal wave-one pattern in equatorial tropospheric ozone. The column-integrated tropospheric ozone amount is greater over the Atlantic stations than over the Pacific and eastern Indian Ocean. This is because the upper troposphere over Watukosek (for example) shows the imprint of convection, with lower ozone injected upward whereas the upper troposphere over Ascension or Natal are enriched by ozone over 65 ppbv. The result is a standing wave pattern, seen in longitudinal cross-section in Figure 7.4-5. The characterization of the wave-one through the ozone sounding record resolved a decade of argument initiated by Shiotani's discovery of the wave-one [Shiotani, 1992; Shiotani and Hasebe, 1994; Newchurch et al., 2001]. Analysis of satellite data, with large instrumental uncertainties in the lower stratosphere analysis, is not sufficient to discriminate between a stratospheric or tropospheric feature. The vertical resolution of the sounding record makes definitive identification of the wave feature possible [Thompson et al., 2003].

Although tropospheric ozone transport from Africa toward South America is prevalent, analysis of air parcel origins with back-trajectories during SAFARI-92/TRACE-A showed that in the upper troposphere, the reverse is true. With westerly flow of high ozone (especially post-convective) toward southern Africa and on to the Indian Ocean, high ozone over Irene, for example is sometimes linked to regions of biomass fires over South America (Figure 7.4-6).

7.4.2 Cross-hemispheric Intercontinental Transport and the "Atlantic Paradox"

As pointed out in the previous section, the wave-one pattern, signifying a relatively fixed longitudinal gradient in tropospheric ozone (maximum ozone column over the Atlantic), is mostly southern hemisphere phenomenon. This appears to be due to the Atlantic north of the ITCZ being lower in ozone sources and more affected by convection. A good depiction of the north-south ozone contrast was assembled from TRACE-A aircraft observations by Browell and coworkers [Browell et al., 1996; circles in Figure 7.4-7]. The result is that the highest tropospheric column ozone amount is between 10 and 20S. Satellite observations (Figure 7.4-8) show the north-south contrast is present year-round [Thompson, 2003; Martin et al., 2002]. Note that there is more ozone south of the ITCZ in December-January-February even though biomass fires this time of year lead to regionally high-ozone regions in the northern tropics. As suggested by the annual cycle in tropospheric ozone at the Atlantic sounding stations (Figure 7.4-3), the north-south gradient is most pronounced when south Atlantic subsidence and pyrogenic ozone sources are strongest, in September-October-November (upper panel in Figure 7.4-8)..

The latitudinal cross-section of tropospheric ozone structure over the Atlantic was first determined from ozone soundings launched during oceanographic transects between South America and Europe [Smit et al., 1989]. Weller et al. [1996] noted the relationship between ozone and water vapor on several *Polarstern* cruises, especially low ozone in the upper troposphere in the presence of higher water vapor when convection was present. The January-February 1993 ozonesonde record of Weller et al. [1996] was nearly identical to that measured during the first North America-South Africa cruise ("Aerosols99") with ozonesondes (January-February 1999; Figure 1 in Thompson et al., 2000; Figure 7.4-9). Thompson et al. [2000] designated the appearance of more tropospheric ozone south of the ITCZ when biomass burning was maximizing photochemical ozone production north of the ITCZ as an "ozone paradox" and suggested that the cause was a combination of interhemispheric transport of ozone and greater subsidence and lightning frequency in the southern hemisphere. The contrast in ozone origins (northern Africa vs southern Africa) appears in the air parcel back-trajectories (Figure 7.4-10) that corresponding to the profiles in Figure 7.4-9.

Transport of ozone precursors and other tracers across the ITCZ in the December-January period was observed during the TROPOZ II aircraft experiment (Jonquieres et al., 1998; Jonquieres and Marengo, 1998). Several studies [Edwards et al., 2003; Jenkins et al., 2003; Chatfield et al., 2003] have explored the Atlantic "ozone paradox" with additional data. Edwards et al. [2003] show evidence for cross-hemispheric transport in the satellite CO data (MOPITT) for January 2001. Their mechanism (similar to that in Figure 6a in Thompson, 2003) is shown as the north African-to-Atlantic flow in Figure 7.4-3. Jenkins et al. [2003] and Chatfield et al. [2003] use observations for January 1999 and conclude that lightning and biomass fires both play a role. Chatfield et al [2003] relate a profile over Ascension (3 February 1999) that was taken during the Aerosols99 cruise to lightning and convective redistribution of ozone that occurred during transport from the African continent. In the latter case the ozone layers originating from the burning regions appear above those attributed to lightning (Figure 7.4-11). For a more general interpretation of tropical Atlantic ozone origins, the reader is referred to modeling studies, such as that by Moxim and Levy [2000], Peters et al. [2002] and Martin et al. [2002].

References

- Andreae, M. O., et al., 1994 IGAC book.
- Baldy S., G. Ancellet, M. Bessafi, A. Badr, and D. Lan Sun Luk, Field observations of tropospheric vertical distribution of tropical ozone at a remote marine site in the southern hemisphere, *J. Geophys. Res.*, **101**, 23,835-23,849, 1996.
- Browell, E. V., et al., JGR, 1996.
- Cahoon, D. R., Jr., B. J. Stocks, J. S. Levine, W. R. Cofer III and K. P. O'Neill, Seasonal distribution of African savanna fires, *Nature*, **359**, 812-815, 1992.
- Chatfield, R. B., and A. C. Delany, Convection links biomass burning to increased tropical ozone: However, models will tend to overpredict O₃, *J. Geophys. Res.*, **95**, 18473-18488, 1990.
- Chatfield, R. B., J. A. Vastano, H. B. Singh, and G. W. Sachse, A general model of how fire emissions and chemistry produce African/Oceanic plumes (O₃, CO, PAN, smoke) seen in TRACE-A, *J. Geophys. Res.*, **101**, 24,279-24,306, 1996.
- Chatfield, R. B., Z. Guo, G. W. Sachse, D. R. Blake and N. J. Blake, The subtropical global plume in the Pacific Exploratory Mission-Tropics A (PEM-Tropics A), PEM-Tropics B, and the Global Atmospheric Sampling Program (GASP): How tropical emissions affect the remote Pacific, *J. Geophys. Res.*, **107**, D16, doi: 10.129/2001JD000497, 2002.
- Chatfield, R. B., H. Guan, A. M. Thompson, R. D. Hudson, J. C. Witte, Lightning and other influences on Tropical Tropospheric Ozone: Empirical studies of covariance, Paper presented at the IUGG, 8 July 2003, Sapporo, Japan.
- Connors, V. S., et al., 1999.
- Cosijn and Tyson, 1996
- Cros et al., 1988.
- Crutzen et al., 1979
- Crutzen and Seiler, 1980. or Seiler and Crutzen, 1980??
- Crutzen et al., 1985
- de Laat and Lelieveld, 2002.
- Delany et al., 1985
- Diab, R. D., A. Raghunandran, A. M. Thompson, V. Thouret, Classification of tropospheric ozone profiles over Johannesburg based on MOZAIC aircraft data, *Atmos. Chem. Phys.*, in press, 2003.
- Duncan, B., et al.,
- Edwards, D. P.,
- Fenn M. A., et al., Ozone and aerosol distributions and air mass characteristics over the South Pacific during the burning season, *J. Geophys. Res.* **104**, 16197-16212, 1999.
- Fishman, J., C. E. Watson, J. C. Larsen, and J. A. Logan, The distribution of tropospheric ozone determined from satellite data, *J. Geophys. Res.*, **95**, 3599-3617, 1990.
- Fishman, J., et al., Use of satellite data to study tropospheric ozone in the tropics, *Science.*, **xx**, 1991.
- Folkins, I., ??C. Braun, A. M. Thompson, J. C. Witte, Tropical ozone as in indicator of deep convective outflow, *J. Geophys. Res.*, **107**, D13, doi: 10.129/2001JD001178, 2002.
- Fujiwara, M., K. Kita, S. Kawakami, T. Ogawa, N. Komala, S. Saraspriya, and A. Surtipriya, Tropospheric ozone enhancements during the Indonesian forest fire events in 1994 and in 1997 as revealed by ground-based operations, *Geophys. Res. Lett.*, **26**, 2,147-2,420,

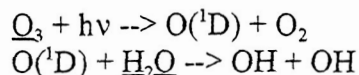
- 1999.
- Fujiwara, M., K. Kita, T. Ogawa, S. Kawakami, T. Sano, N. Komala, S. Saraspriya, A. Suropto, Seasonal variation of the tropospheric ozone in Indonesia revealed by 5-year ground based observations, *J. Geophys. Res.*, **105**, 1879-1888, 2000.
- Garstang M., P. D. Tyson, R. J. Swap, M. Edwards, P. Kållberg, and J. A. Lindesay, Horizontal and vertical transport of air over southern Africa, *J. Geophys. Res.*, **101**, 23,721-23,736, 1996.
- Gregory, G L 1999
- Hauglataine ????
- Hoell J. M., D. D. Davis, D. J. Jacob, M. O. Rodgers, R. E. Newell, H. E. Fuelberg, R. J. McNeal, J. L. Raper and R. J. Bendura, Pacific Exploratory Mission in the tropical Pacific: PEM-Tropics A, August-September 1996, *J. Geophys. Res.*, **104**, 5567-5583, 1999.
- Hudson, R. D., and A. M. Thompson, Tropical tropospheric ozone (TTO) from TOMS by a modified-residual method, *J. Geophys. Res.*, **103**, 22129-22145, 1998.
- Jacob, D. J., et al., Origin of ozone and NO_x in the tropical troposphere: A photochemical analysis of aircraft observations over the South Atlantic Basin, *J. Geophys. Res.*, **101**, 24,235-24,250, 1996.
- Jenkins, G. R., J-H. Ryu, A. M. Thompson, J. C. Witte, Linking horizontal and vertical transport of biomass fire emissions to the tropical Atlantic ozone paradox during the northern hemisphere winter season. II. 1998-1999, *J. Geophys. Res.*, in press, 2003.
- Jonquière, I., A. Marengo, A. Maalej and F. Rohrer, Study of ozone formation and transatlantic transport from biomass burning emissions over West Africa during the airborne Tropospheric Ozone Campaigns TROPOZ I and TROPOZ II, *J. Geophys. Res.*, **103**, 19,059-19,073, 1998.
- Jonquière, I., and A. Marengo, Redistribution by deep convection and long-range transport of CO and CH₄ emissions from the Amazon basin, as observed by the airborne campaign TROPOZ II during the wet season, *J. Geophys. Res.*, **103**, 19,075-19,091, 1998.
- Justice, C. O., J. D. Kendall, P. R. Dowty, and R. J. Scholes, Satellite remote sensing of fires during the SAFARI campaign using NOAA-advanced very high resolution radiometer data, *J. Geophys. Res.*, **101**, 23,851-23,863, 1996.
- Kim, J.-H., R. D. Hudson and A. M. Thompson, JGR, 1996.
- Kita et al., 2000 (1997 Indo)
- Kita et al., 2002 (BIBLE)
- Kondo et al., BIBLE overview - 2002a
- Kondo et al., BIBLE paper, 2002b
- Krishnamurti, T. N., M. C. Sinha, M. Kanamitsu, D. Oosterhof, H. Fuelberg, R. Chatfield, D. J. Jacob, and J. Logan, Passive tracer transport relevant to the TRACE-A experiment, *J. Geophys. Res.*, **101**, 23,889-23,907, 1996.
- Lelieveld, J. et al., INDOEX Science summary.
- Levine, Joel S., *Biomass Burning: Atmospheric, Climatic and Biospheric Implications*, MIT Press, Massachusetts, 1991.
- Levine, Joel S., *Biomass Burning and Global Change*, MIT Press, Massachusetts, 1996.
- Logan and Kirchhoff, 1986
- Longo, K. M., et al., Correlation between smoke and tropospheric ozone concentration in Cuiabá during Smoke, Clouds, and Radiation-Brazil (SCAR-B), *J. Geophys. Res.*, **104**, 12,113-

- 12,129, 1999.
- Martin, R. V., 2002
- Matsueda et al., 1999
- Moxim and Levy
- Newchurch, M. J., et al.,
- Newell R. E., V. Thouret, J. Y. N. Cho, P. Stoller, A. Marenco, and H. G. Smit, Ubiquity of quasi-horizontal layers in the troposphere, *Nature*, **398**, 316-319, 1999.
- Olson, J. R., B. A. Baum, D. R. Cahoon, and J. H. Crawford, Frequency and distribution of forest, savanna and crop fires over tropical region during PEM-Tropics A, *J. Geophys. Res.*, **104**, 5865-5876, 1999.
- Oltmans, S. J. et al., Ozone in the Pacific tropical troposphere from ozonesonde observations, *J. Geophys. Res.*, **106**, 32503-32526, 2001.
- S. J. Oltmans, B. J. Johnson, J. M. Harris, A. M. Thompson, H. Y. Liu, H. Vömel, C. Y. Chan, T. Fujimoto, V. G. Brackett, W. L. Chang, J.-P. Chen, J. H. Kim, L. Y. Chan, H.-W. Chang, Tropospheric Ozone over the North Pacific from Ozonesonde Observations, *J. Geophys. Res.*, submitted, 2003. ???
- W. Peters, M. Krol, F. Dentener, A. M. Thompson, J. Lelieveld, Chemistry-transport modeling of the satellite observed distribution of tropical tropospheric ozone, *Atmos. Chem. Phys.*, **2**, 103-120, 2002.
- Peters, W., M. C. Krol, J. P. F. Fortuin, H. M. Kelder, C. R. Becker, A. M. Thompson, J. Lelieveld, P. J. Crutzen, Tropospheric Ozone over a Tropical Atlantic Station in the Northern Hemisphere: Paramaribo, Surinam (6N, 55W), *Tellus B*, in press, 2003.
- Phadnis et al., 2002
- Pickering et al., 1992.
- Pickering K. E., A. M. Thompson, Y. Wang, W-K Tao, D. P. McNamara, V. W. J. H. Kirchhoff, B. G. Heikes, G. W. Sachse, J. D. Bradshaw, G. L. Gregory, and D. R. Blake, Convective transport of biomass burning emissions over Brazil during TRACE-A, *J. Geophys. Res.*, **101**, 23,993-24,012, 1996.
- Piketh
- Ramanathan, ?? INDOEX overview?
- Randriambelo, T., J. L. Baray, S. Baldy, P. Bremaud, and S. Cautenet, A case study of extreme tropospheric ozone contamination in the tropics using in-situ, satellite, and meteorological data, *Geophys. Res. Lett.*, **26**, 1287-1290, 1999.
- Reichle H. G., V. S. Connors, J. A. Holland, R. T. Sherrill, H. A. Wallio, J. C. Casas, E. P. Condon, B. B. Gormsen and W. Seiler, The distribution of middle tropospheric carbon-monoxide during early October 1984, *J. Geophys. Res.*, **95**, 9845-9856, 1990.
- Schultz, M. G., et al., On the origins of tropospheric ozone and NO_x over the tropical South Pacific, *J. Geophys. Res.*, **104**, 5829-5844, 1999.
- Shiotani, M., 1992.
- Shiotani and Hasebe, 1994
- Smit, H. G. J., 1989.
- Smyth S. B., et al., Factors influencing the upper free tropospheric distribution of reactive nitrogen over the South Atlantic during the TRACE A experiment, *J. Geophys. Res.*, **101**, 24165-24186, 1996.
- Stoller, P., et al., Measurements of atmospheric layers from the NASA DC-8 and P3-B aircraft during PEM-Tropics A, *J. Geophys. Res.*, **104**, 5745-5764, 1999.

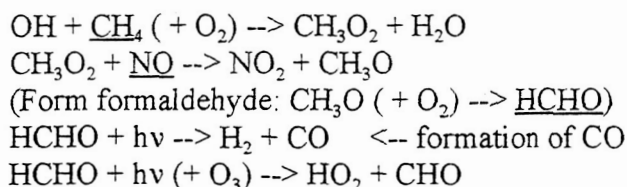
- R. J. Swap, H. J. Annegarn, J. T. Suttles, J. Haywood, M. C. Hemmlinger, C. Hely, P. V. Hobbs, B. N. Holben, J. Ji, M. D. King, T. Landmann, W. Maenhaut, L. Otter, B. Pak, S. J. Piketh, S. Platnick, J. Privette, D. Roy, A. M. Thompson, D. Ward, R. Yokelson, The Southern African Regional Science Initiative (SAFARI-2000): Dry-Season Campaign, an Overview, *S. Afr. J. Science*, **98**, 125-130, 2002.
- Taupin
- Thompson, A. M., "Biomass Burning," Chapter 14 in *Handbook of Weather, Climate, and Water: Atmospheric Chemistry, Hydrology, and Societal Impacts*, ed. T. D. Potter and B. R. Colman, J. Wiley and Sons, 2003.
- Thompson, A. M. and R. D. Hudson, Tropical Tropospheric Ozone (TTO) Maps from Nimbus 7 and Earth-Probe TOMS by the Modified-Residual Method: Evaluation, El Niño Signals and Trends based on Atlantic Regional Time Series, *J. Geophys. Res.*, **26**, 961-26,975, 1999.
- Thompson, A. M., K. E. Pickering, D. P. McNamara, M. R. Schoeberl, R. D. Hudson, J. H. Kim, E. V. Browell, V. W. J. H. Kirchhoff, and D. Nganga, Where did tropospheric ozone over southern Africa and the tropical Atlantic come from in October 1992? Insights from TOMS, GTE/TRACE-A and SAFARI-92, *J. Geophys. Res.*, **101**, 24,251-24,278, 1996.
- Thompson, A. M., B. G. Doddridge, J. C. Witte, R. D. Hudson, W. T. Luke, J. E. Johnson, B. J. Johnson, S. J. Oltmans, and R. Weller, A tropical Atlantic ozone paradox: Shipboard and satellite views of a tropospheric ozone maximum and wave-one in January-February 1999, *Geophys. Res. Lett.*, **27**, 3317-3320, 2000.
- Thompson, A. M., J. C. Witte, R. D. Hudson, H. Guo, J. R. Herman, M. Fujiwara, Tropical tropospheric ozone and biomass burning, *Science*, **291**, 2128-2132, 2001.
- Thompson, A. M., J. C. Witte, M. T. Freiman, N. A. Phahlane, G. J. R. Coetzee, Lusaka, Zambia, during SAFARI-2000: Convergence of Local and Imported Ozone Pollution, *Geophys. Res. Lett.*, **29**, 1976, doi: 10.129/2002GL015399, 2002.
- Thompson, A. M., J. C. Witte, S. J. Oltmans, F. J. Schmidlin, J. A. Logan, M. Fujiwara, V. W. J. H. Kirchhoff, F. Posny, G. J. R. Coetzee, B. Hoegger, S. Kawakami, T. Ogawa, J. P. F. Fortuin, H. M. Kelder, Southern Hemisphere ADditional Ozonesondes (SHADOZ) 1998-2000 tropical ozone climatology. 2. Tropospheric Variability and the Zonal Wave-One, *J. Geophys. Res.*, **108**, 8241, doi: 10.129/2002JD002241, 2003.
- Tyson, P. D., M. Garstang, A. M. Thompson, P. D'Abreton, R. D. Diab, E. V. Browell, Atmospheric transport and photochemistry of ozone over central Southern Africa during the Southern Africa Fire-Atmosphere Research Initiative, *J. Geophys. Res.*, **102**, 10,623-10,635, 1997.
- van Wilgen, B. W., M. O. Andreae, J. G. Goldammer, and J. A. Lindesay, *Fire in the Southern Africa Savannas: Ecological and Atmospheric Perspectives*, Witwatersand University Press, Johannesburg, South Africa, 1997.
- Weller, R., R. Lilischkis, O. Schrems, R. Neuber, and S. Wessel, Vertical ozone distribution in the marine atmosphere over the central Atlantic Ocean (56°S-50°N), *J. Geophys. Res.*, **101**, 1387-1399, 1996.

Table 7-1. Photochemical reactions linking methane, NMHC, CO and NO with Ozone

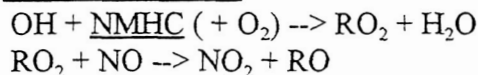
OH Forms from Ozone Photolysis:



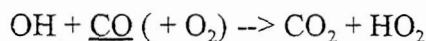
Methane Oxidation:



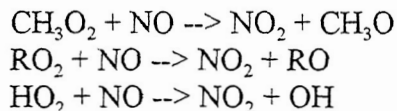
NMHC Oxidation:



CO Oxidation by OH produces HO₂:



Conversion of NO to NO₂ by HO₂, RO₂, CH₃O₂:



Formation of O₃:

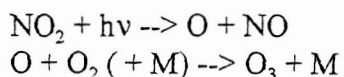


Table 7-2. Campaigns in which ozone and biomass burning transport were investigated

Name and Date

BIBLE - Biomass Burning and Lightning Experiment 1998, 1999
 DECAFE - Dynamique et Chimie Atmosphere des Forets Equatoriale 1988
 EXPRESSO
 FOS/DECAFE - Fires in Savannas - 1991
 INDOEX - Indian Ocean Experiment - 1999
 LBA/CLAIRE - Large-Scale Biosphere-Atmosphere
 SAFARI-1992 - Southern African Fire Atmospheric Research Initiative
 SAFARI-2000- Southern African Atmospheric Research Initiative
 STRAT0Z
 TRACE-A - Transport and Atmospheric Chemistry near the Equator- Atlantic - 1992

TROPOZ

=====

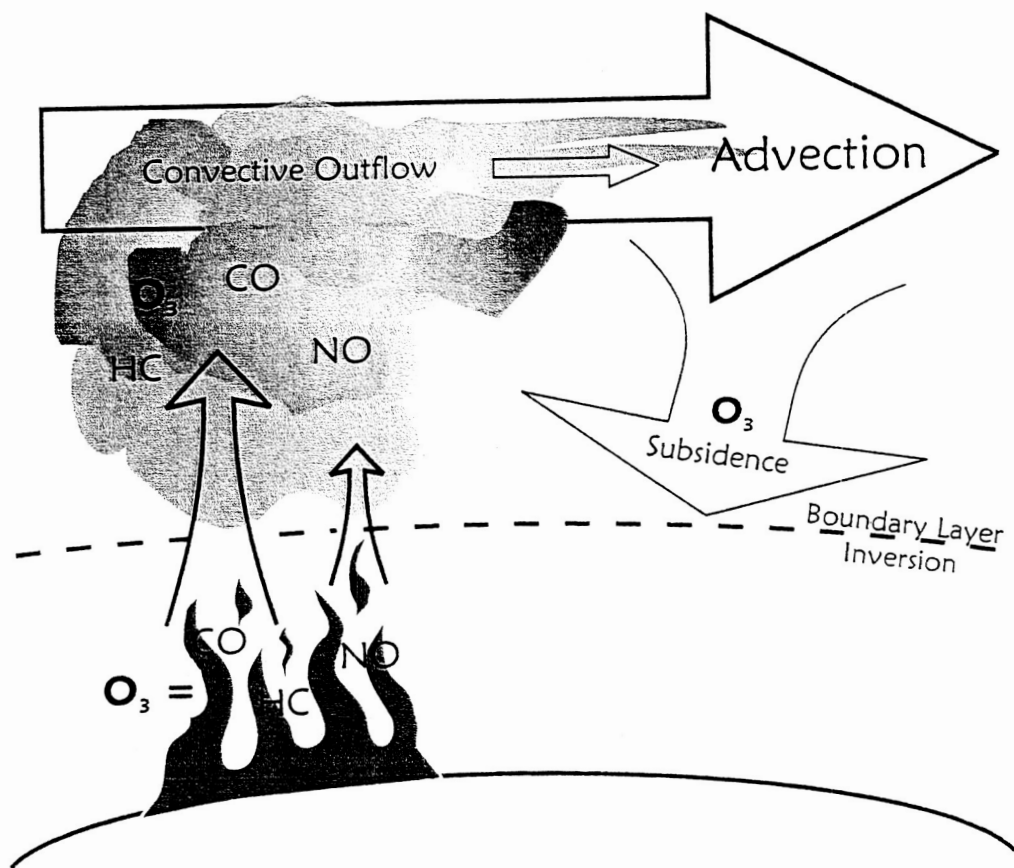


Fig. 7.1-1

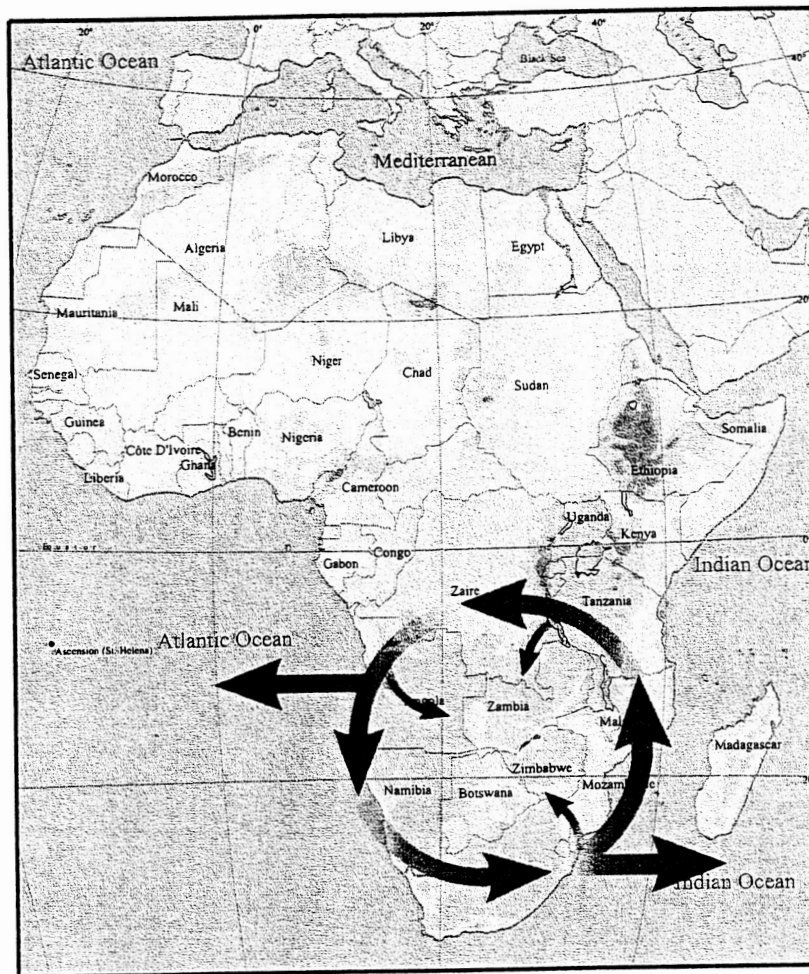


Fig. 7.1-2

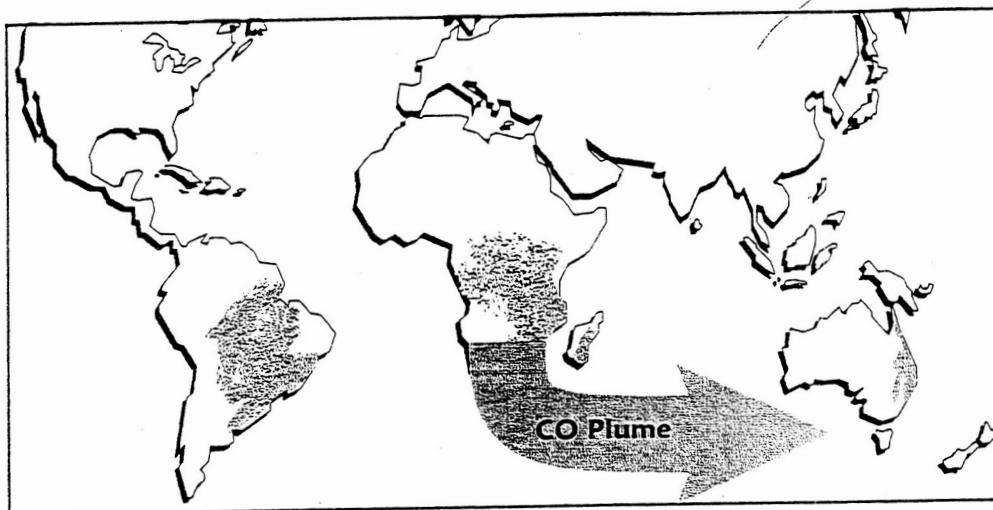


Fig. 7.2-1

La Reunion 1996 Profiles

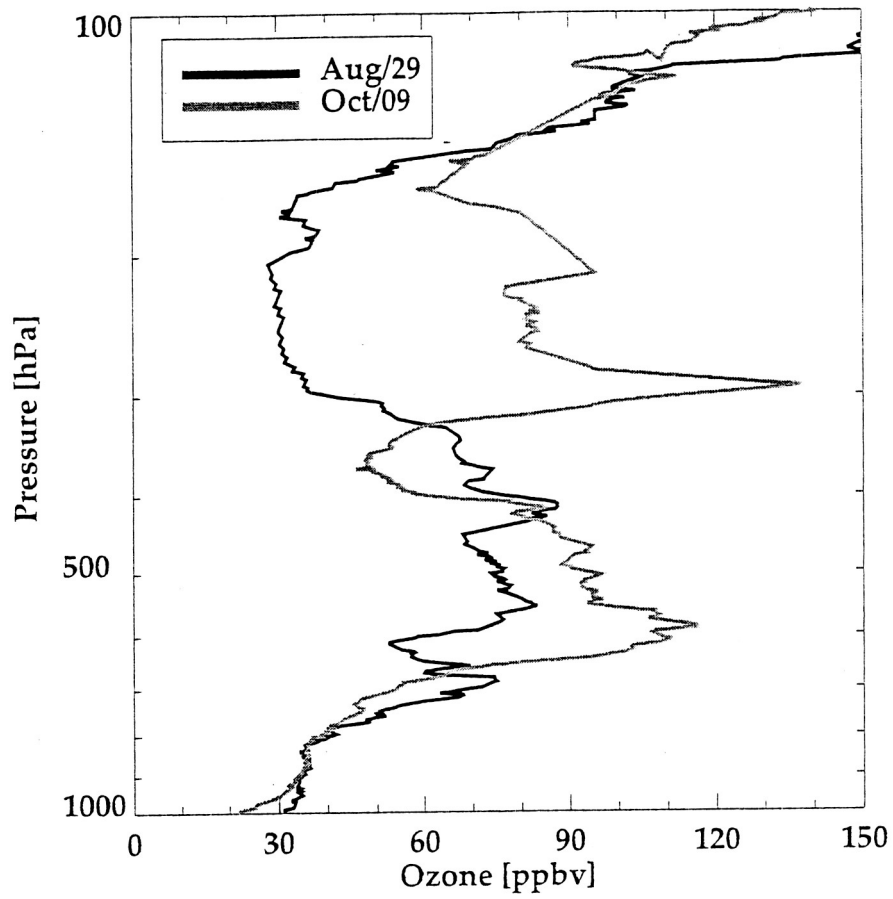


Fig. 7.2-2

8-Day Back-trajectories over La Reunion (21S,56E)

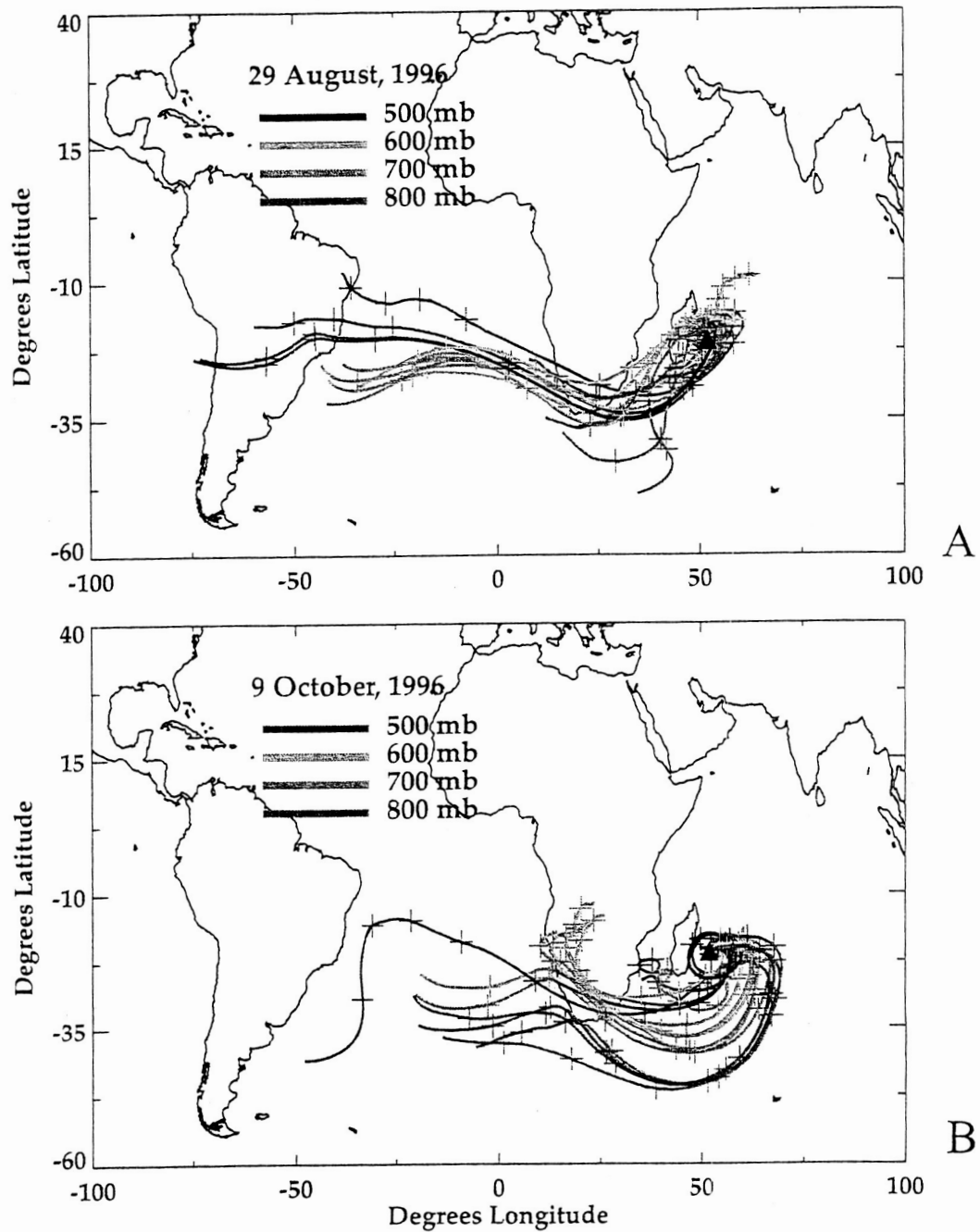


Fig. 7.2-3

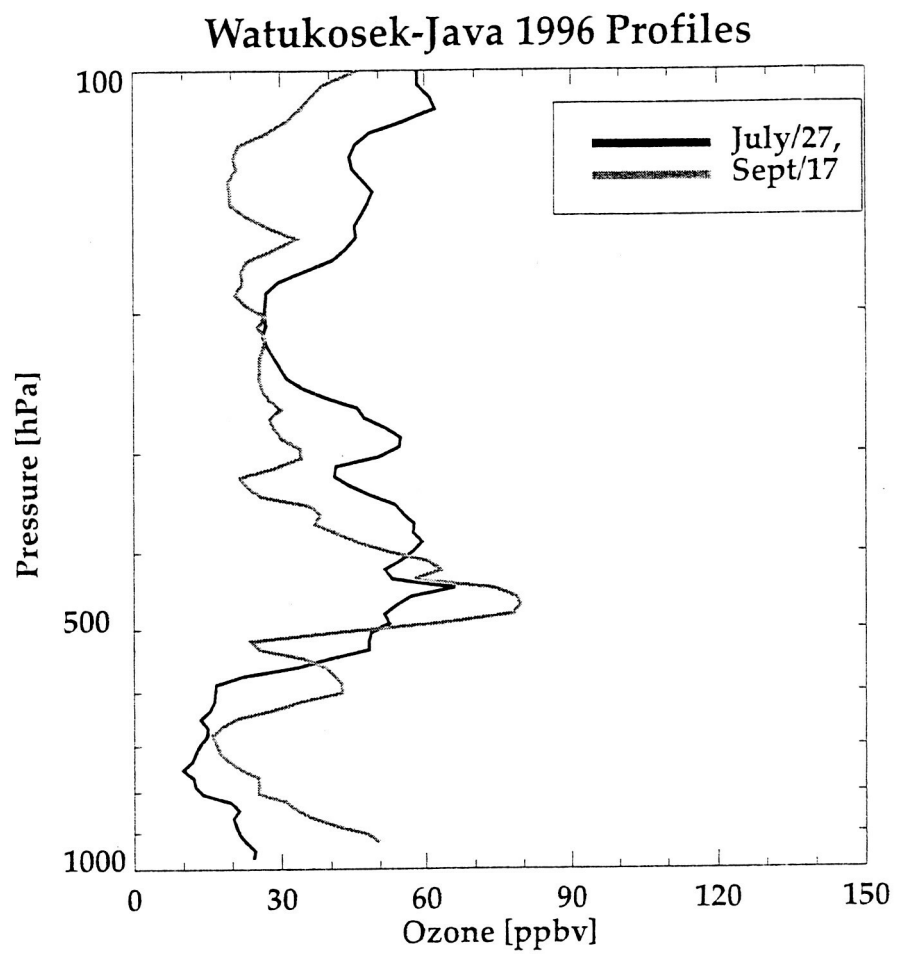


Fig. 7.2-4

8-Day Back-trajectories over Watukosek-Java (8S,113E)

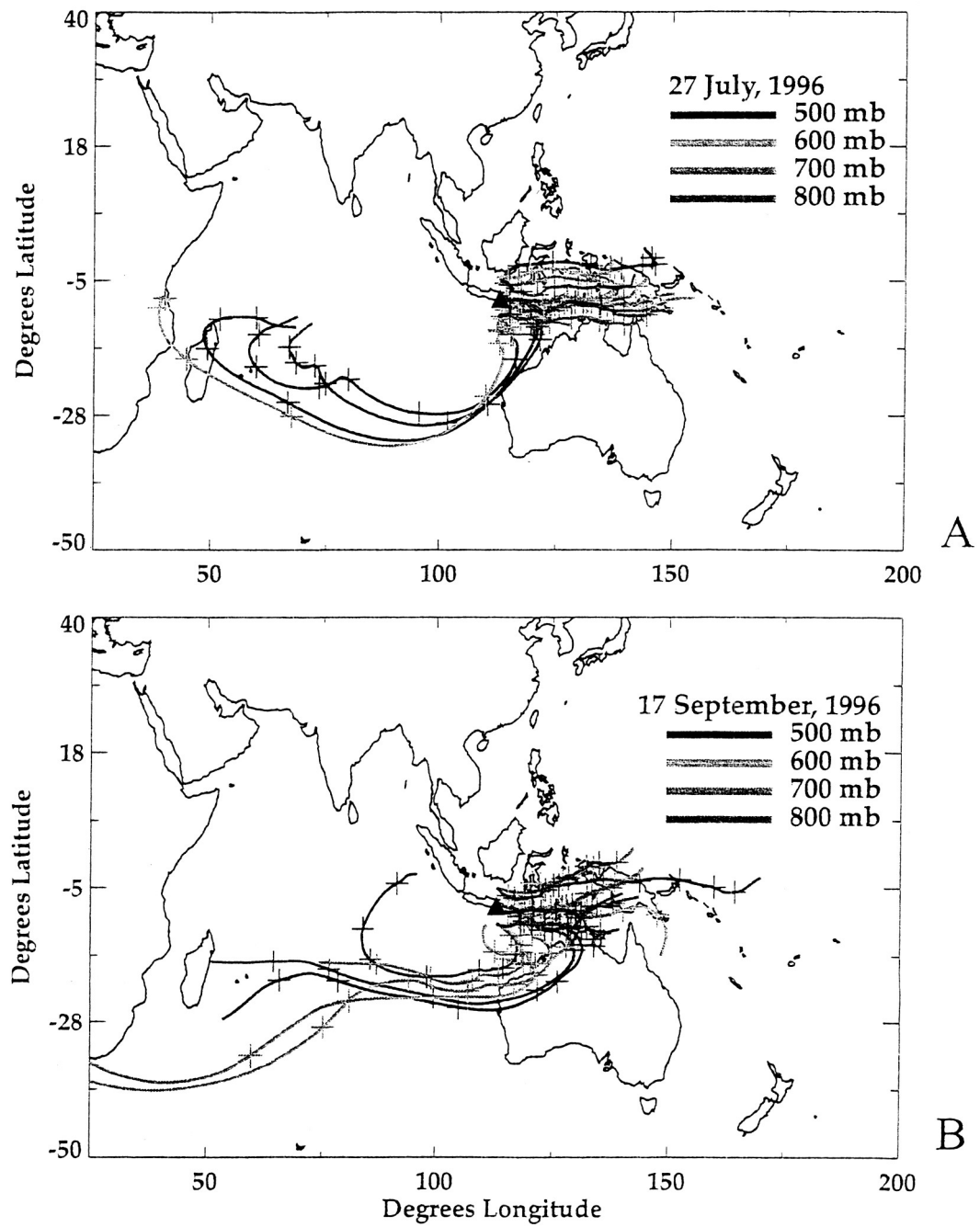


Fig. 7.2-5

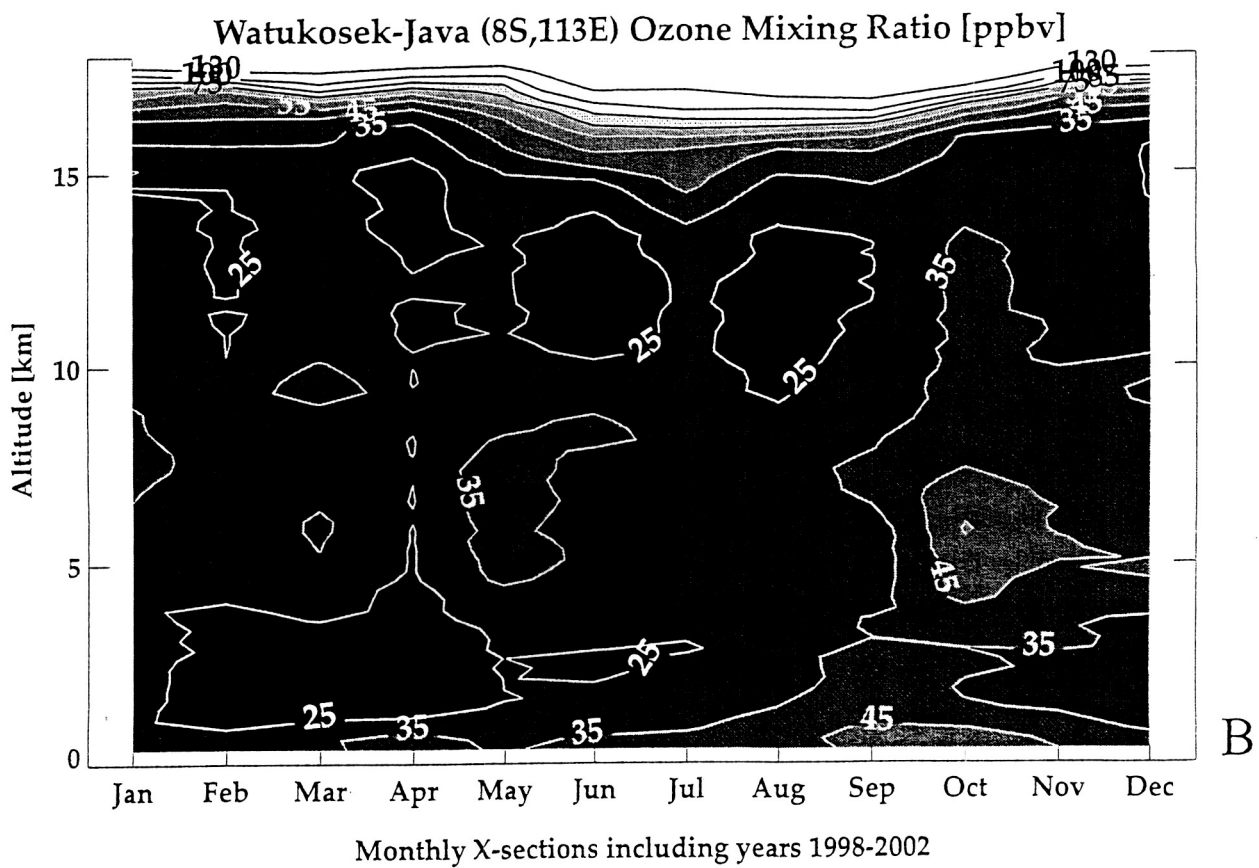
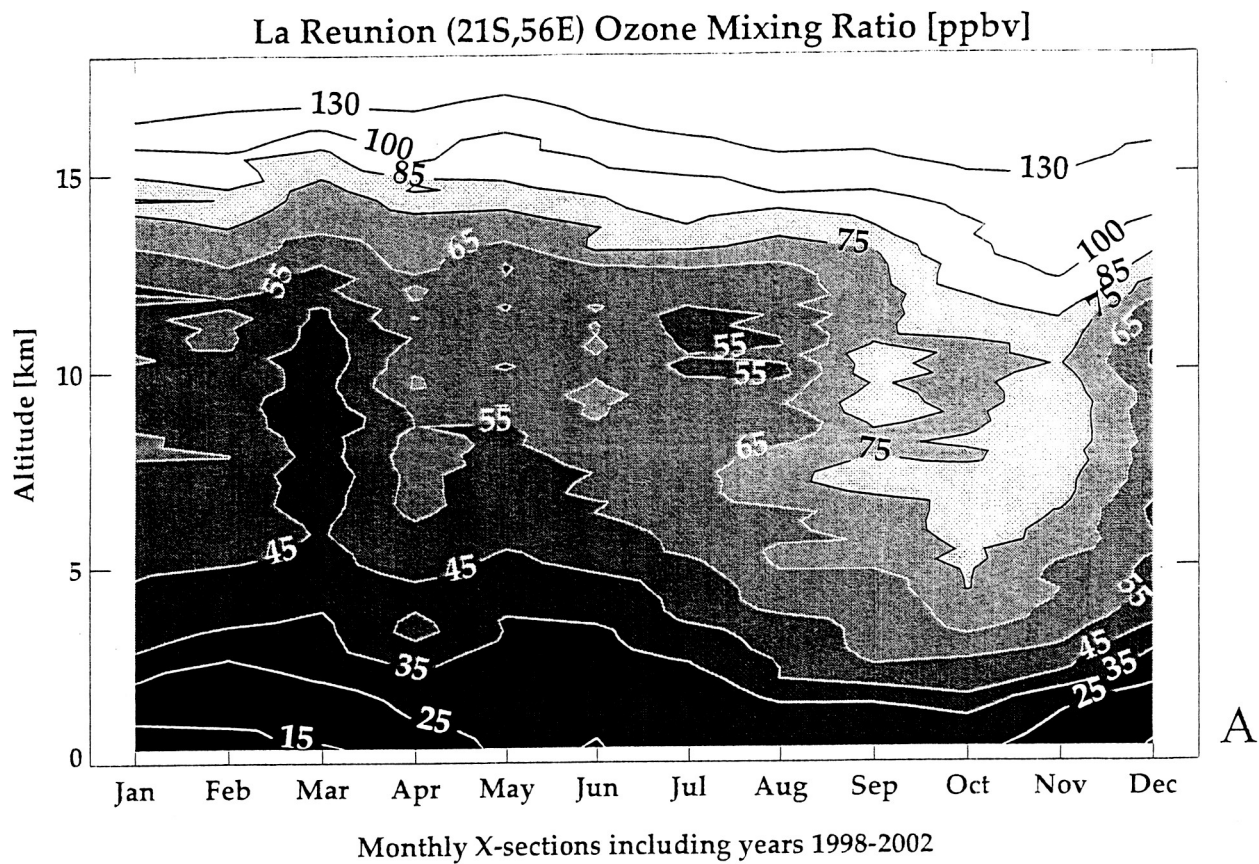


Fig. 7.2-6

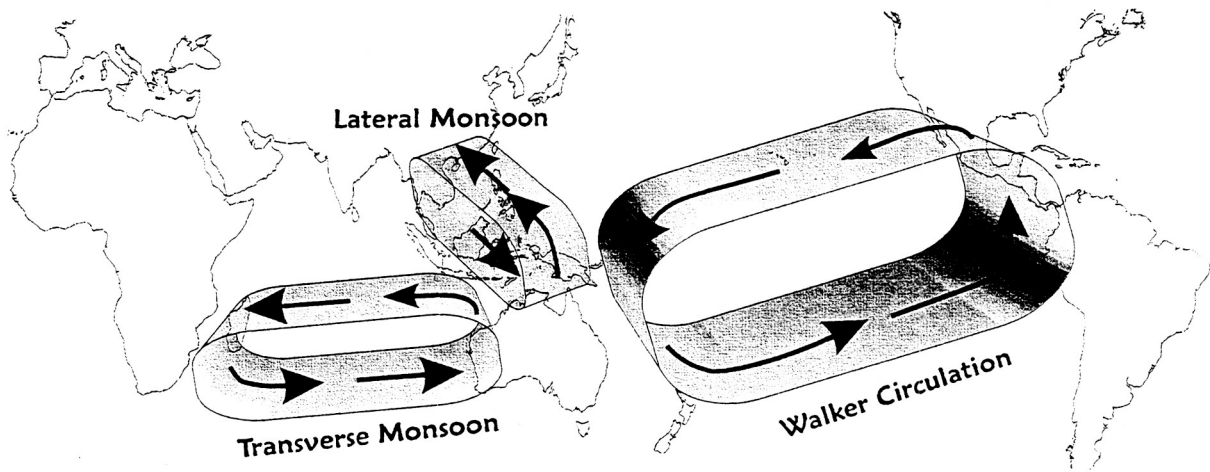


Fig. 7.2-7

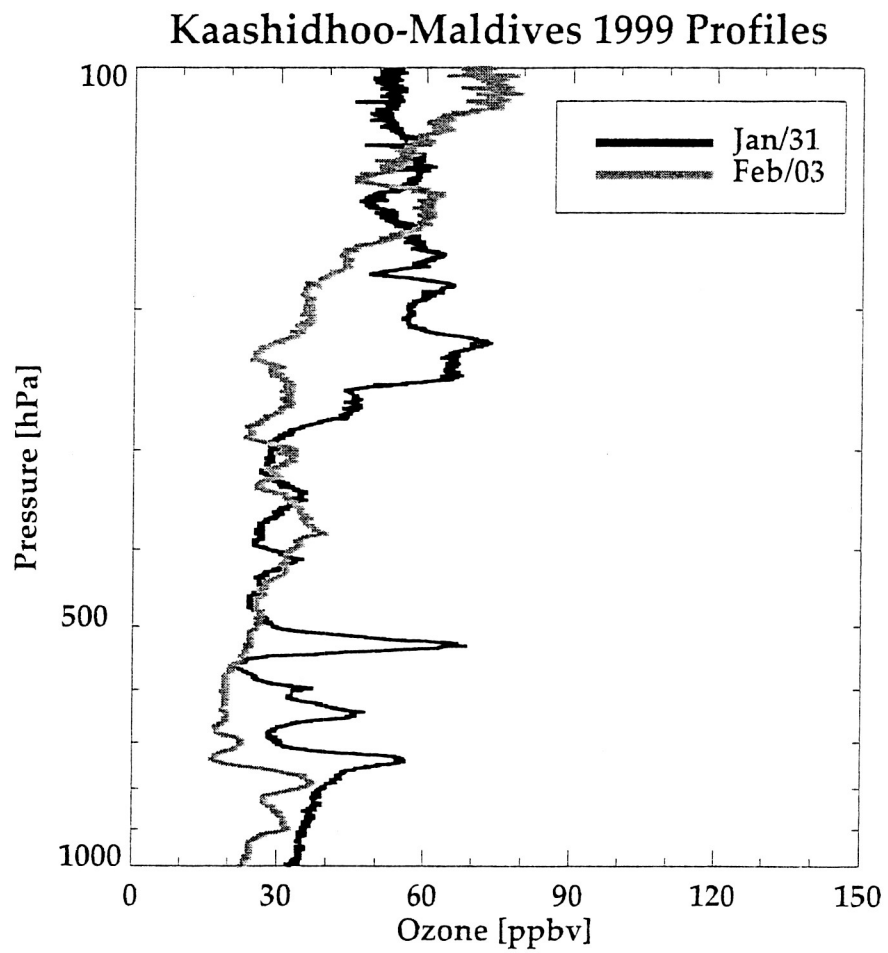


Fig. 7.2-8

8-Day Back-trajectories over Kaashidhoo (5N,74E)

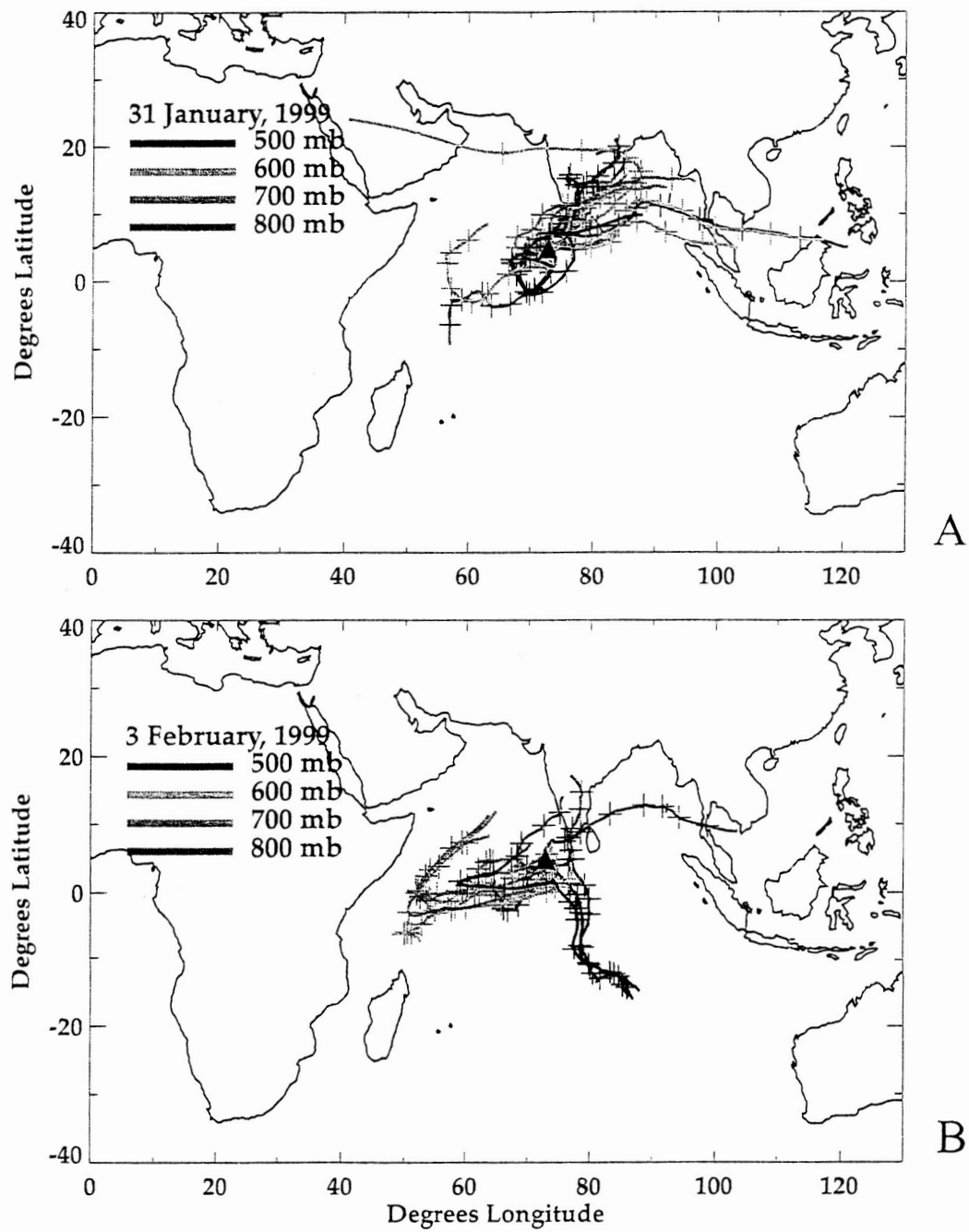


Fig. 7.2-9

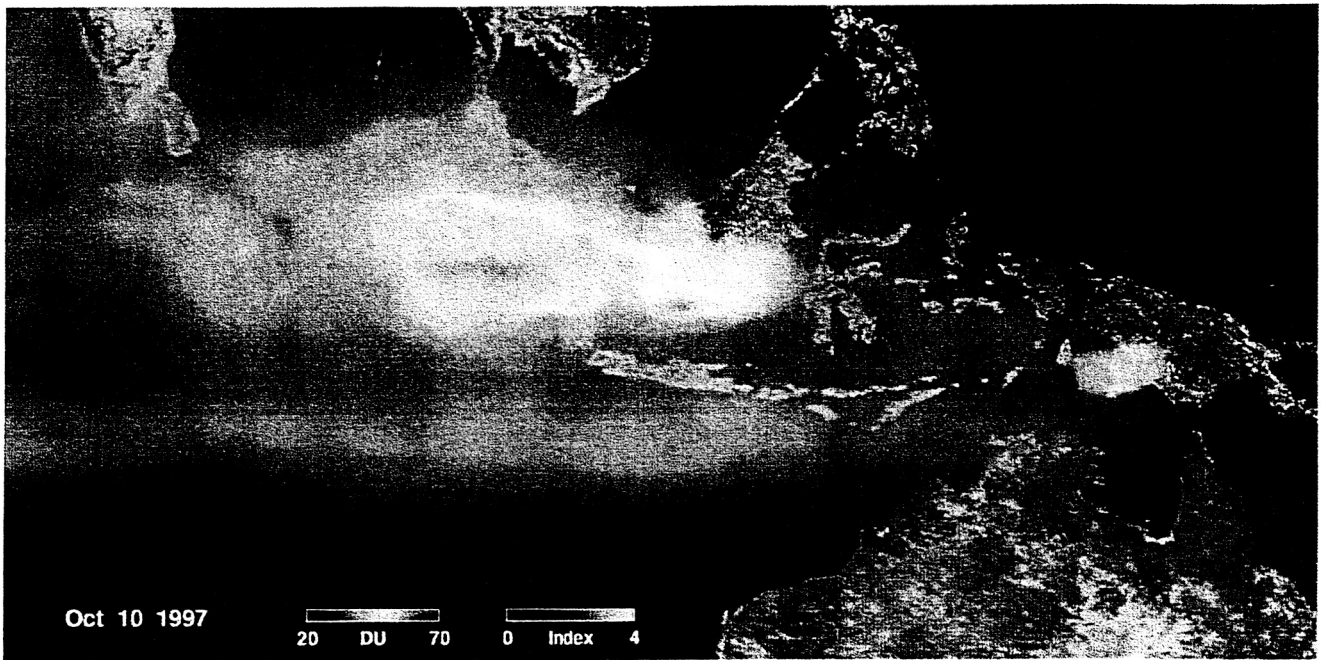


Fig. 7.2-10

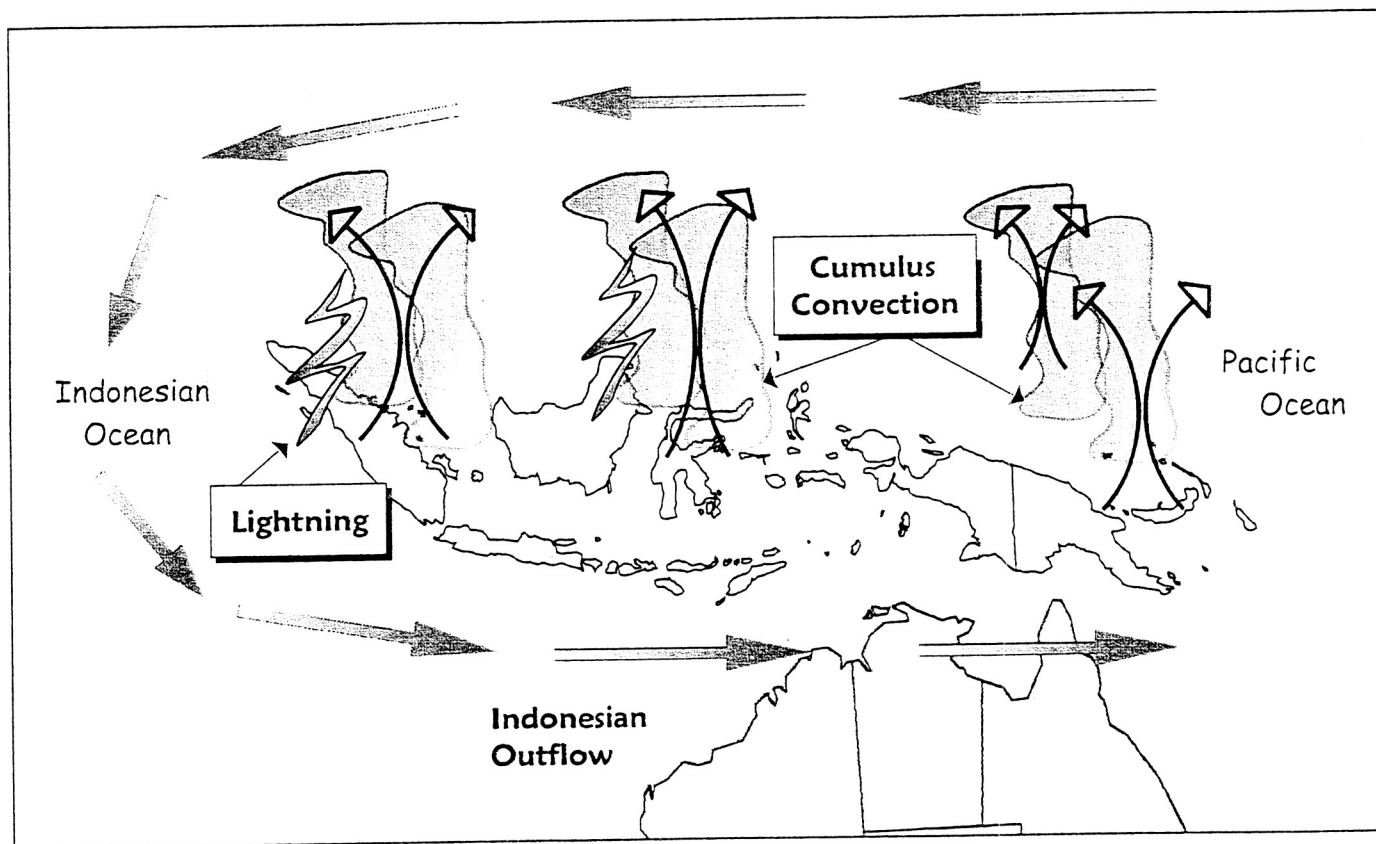
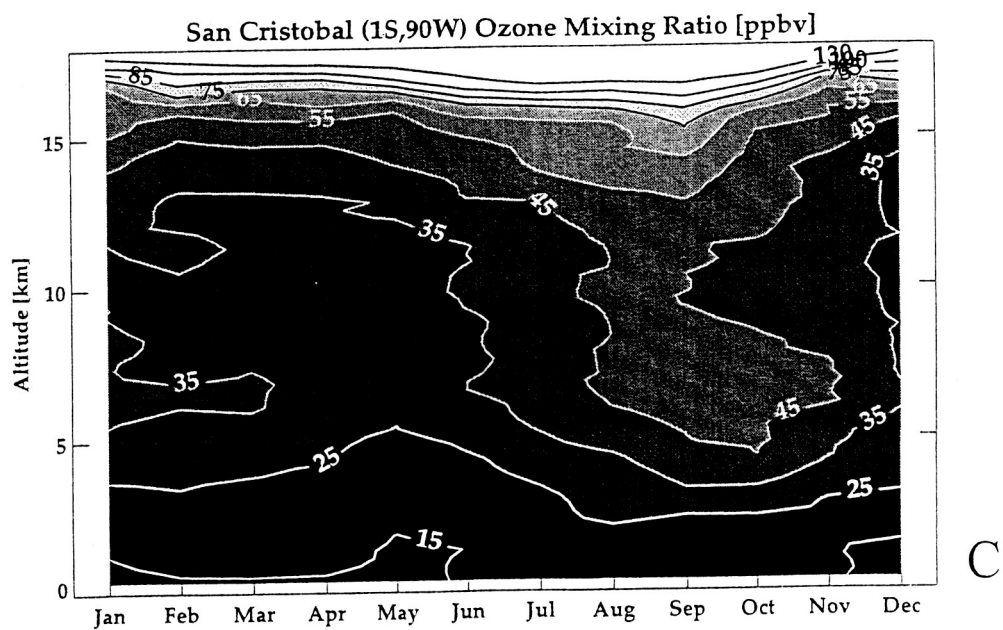
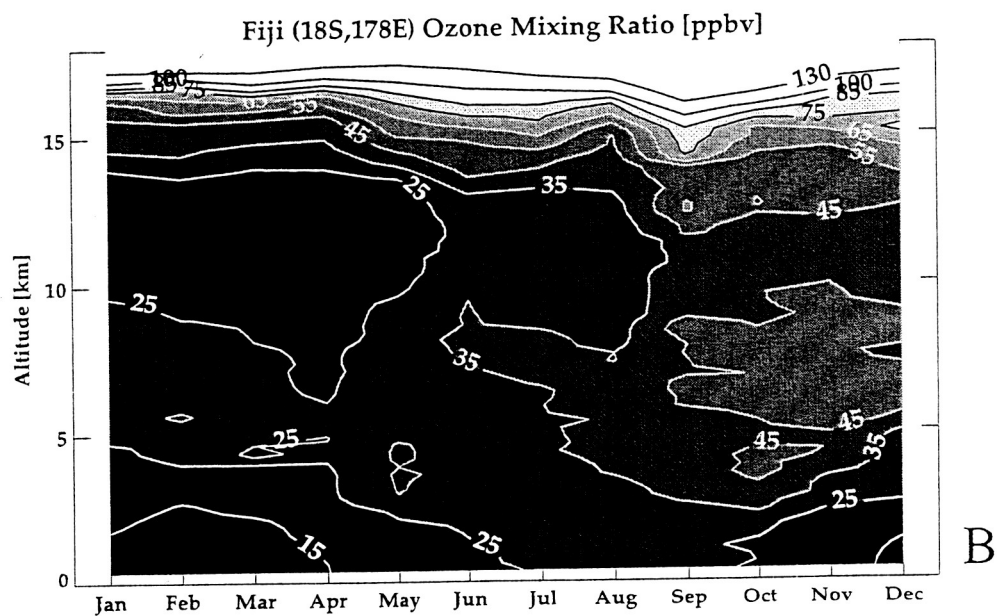
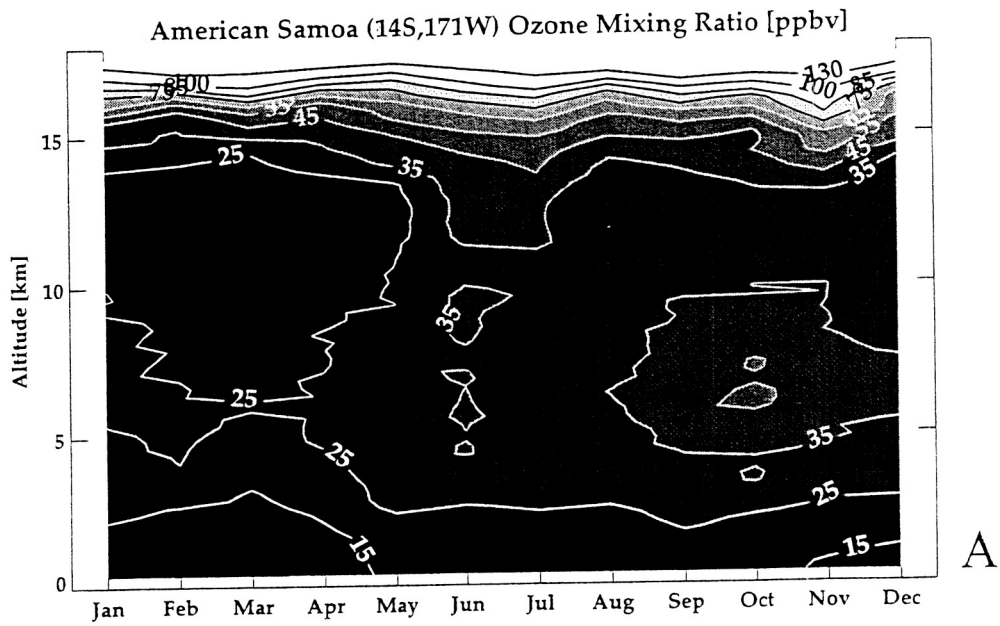
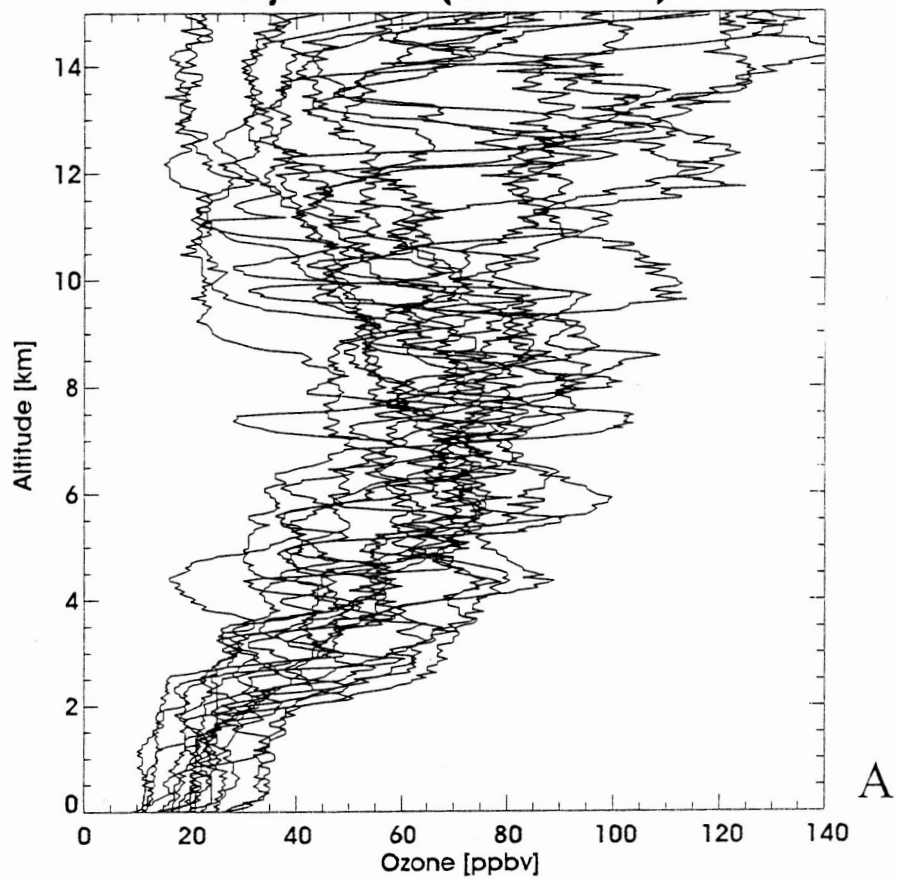


Fig. 7.2-11



Fiji Profiles (1998-2002)



American Samoa Profiles (1998-2002)

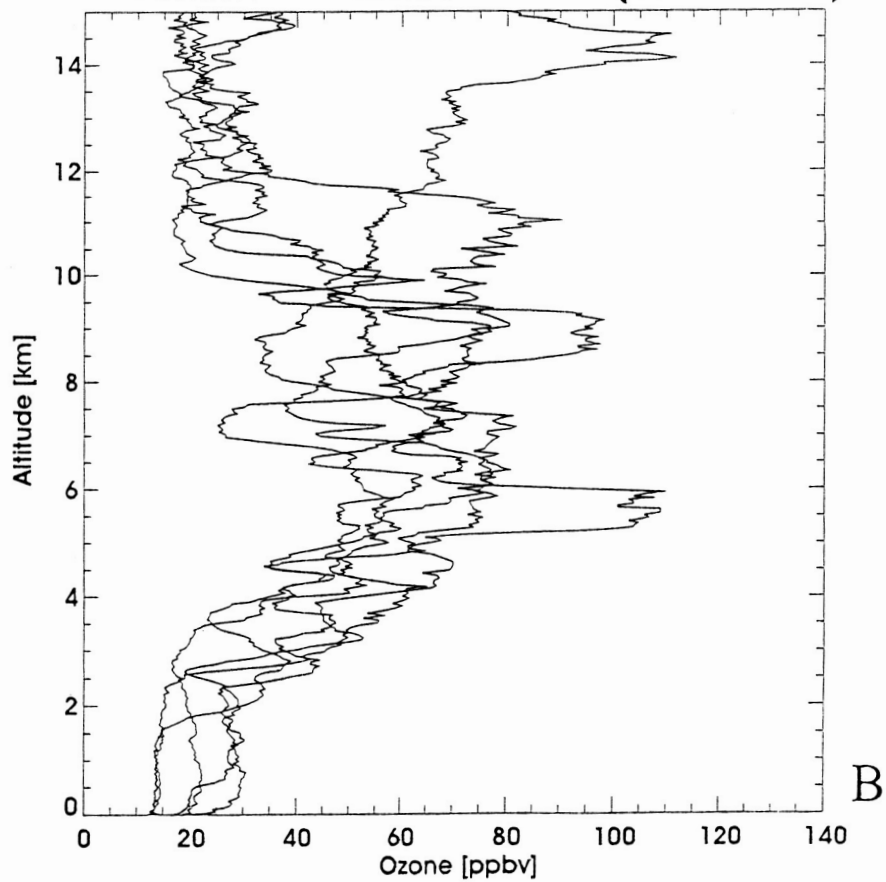


Fig. 7.3-2

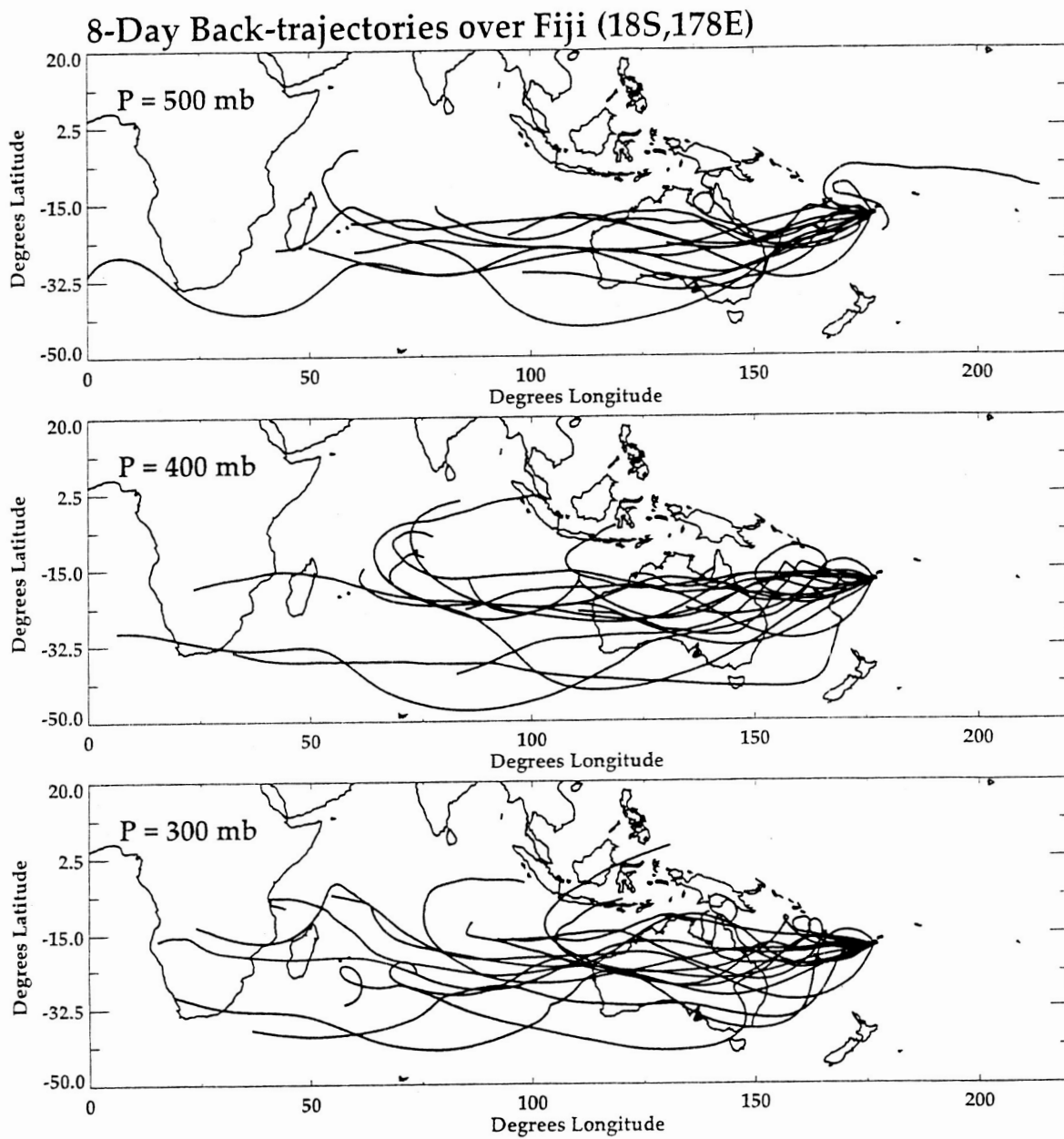


Fig. 7.3-3A

8-Day Back-trajectories over American Samoa (14S,171W)

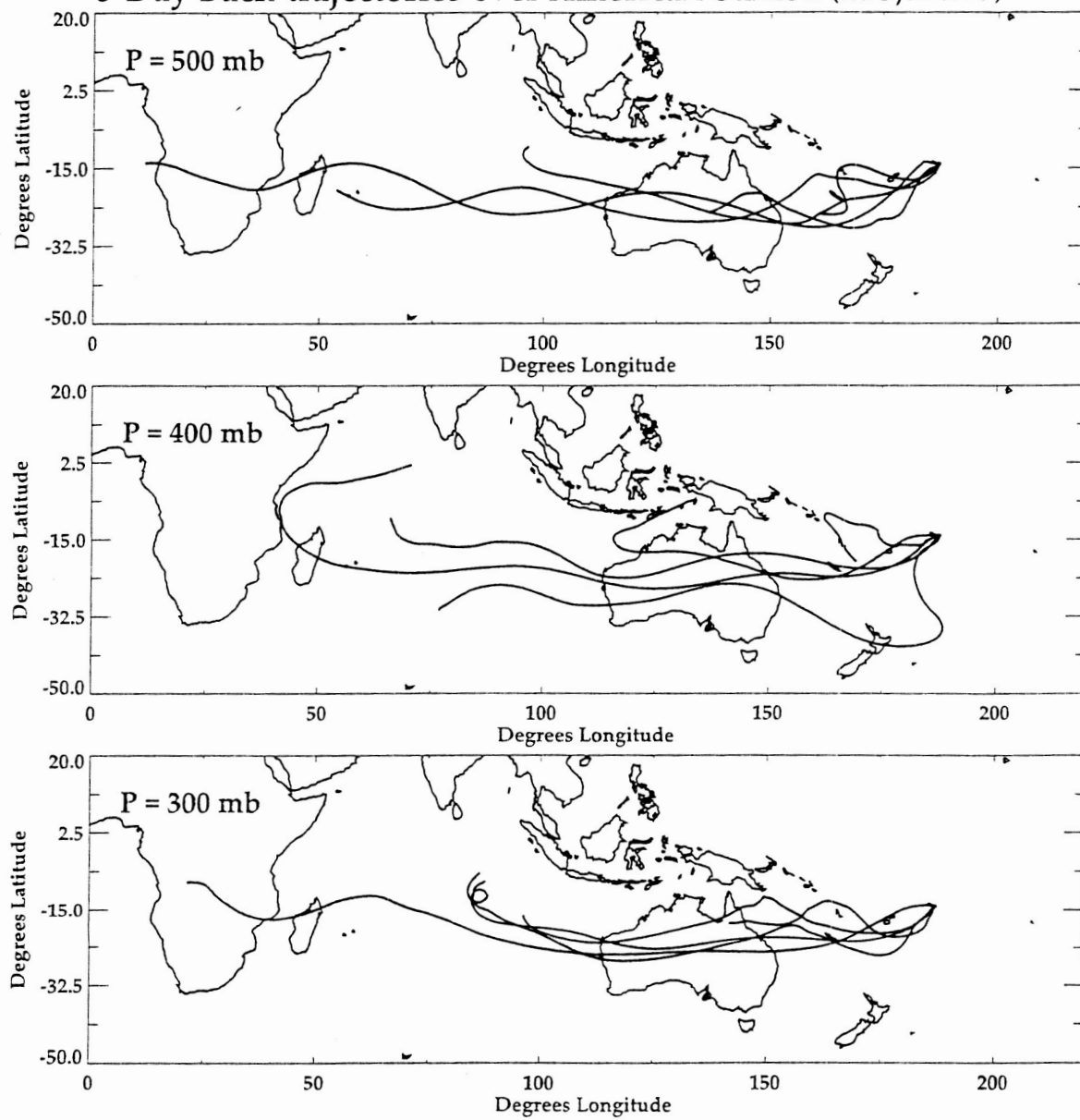


Fig. 7.3-3B

San Cristobal Profiles (1998-2002)

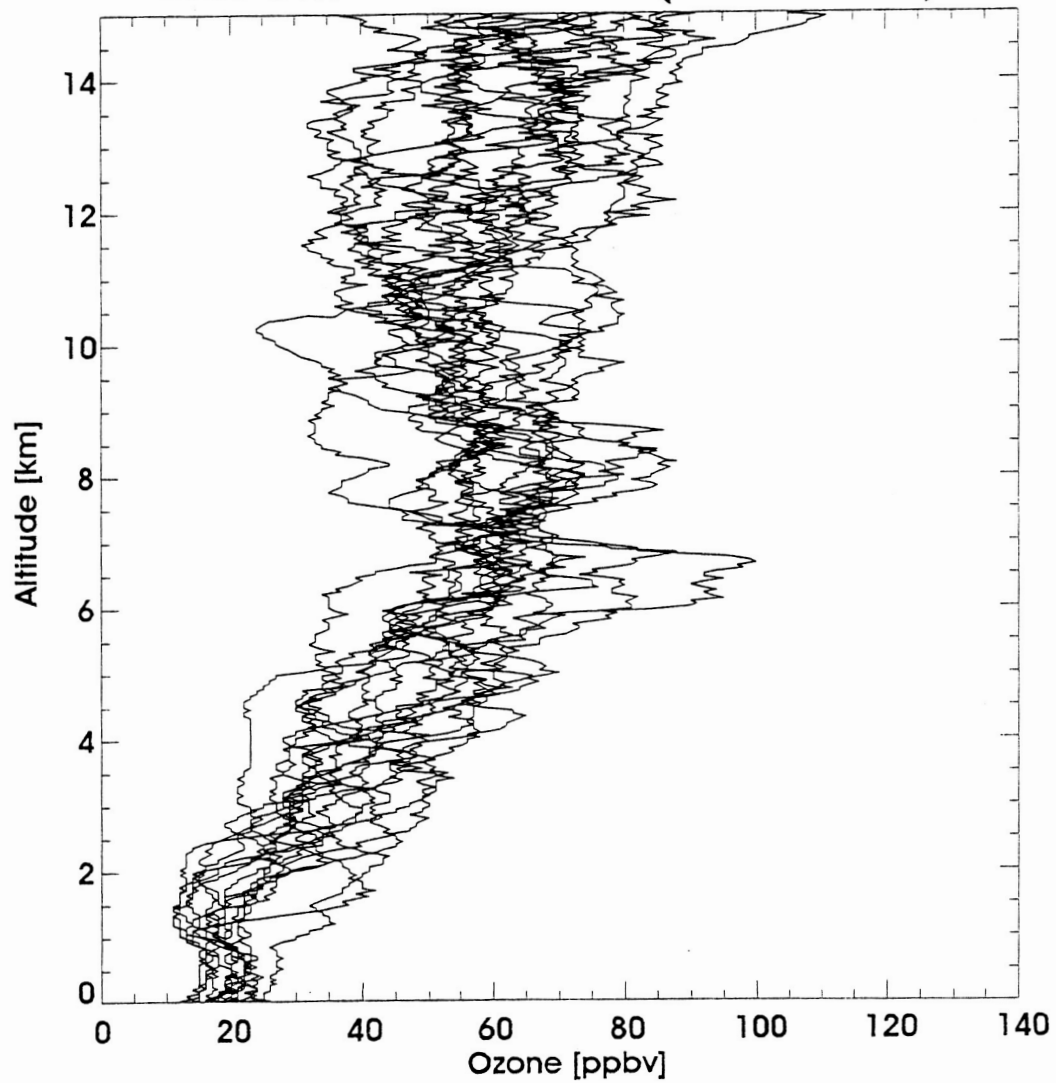


Fig. 7.3-4A

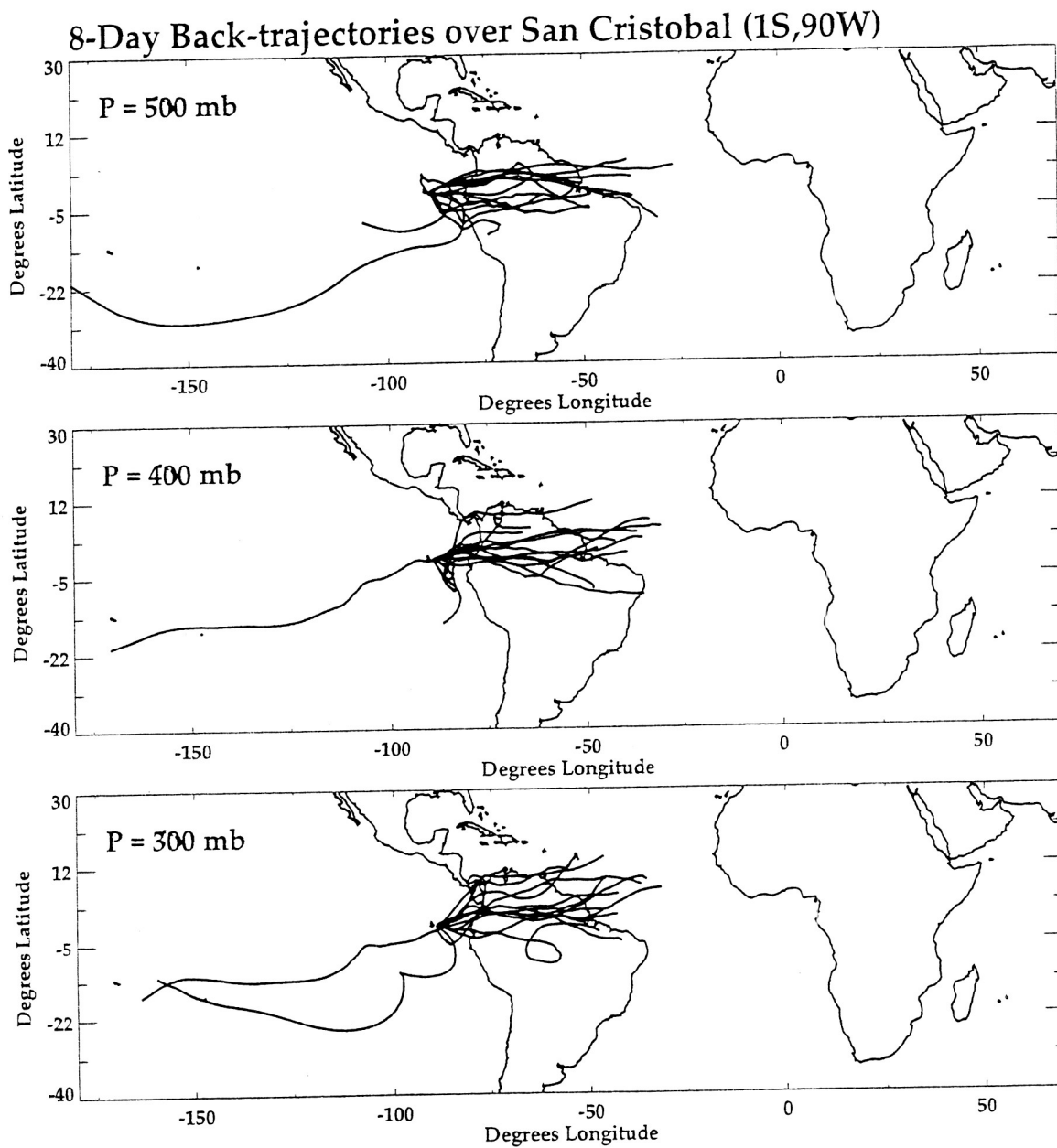


Fig. 7.3-4B

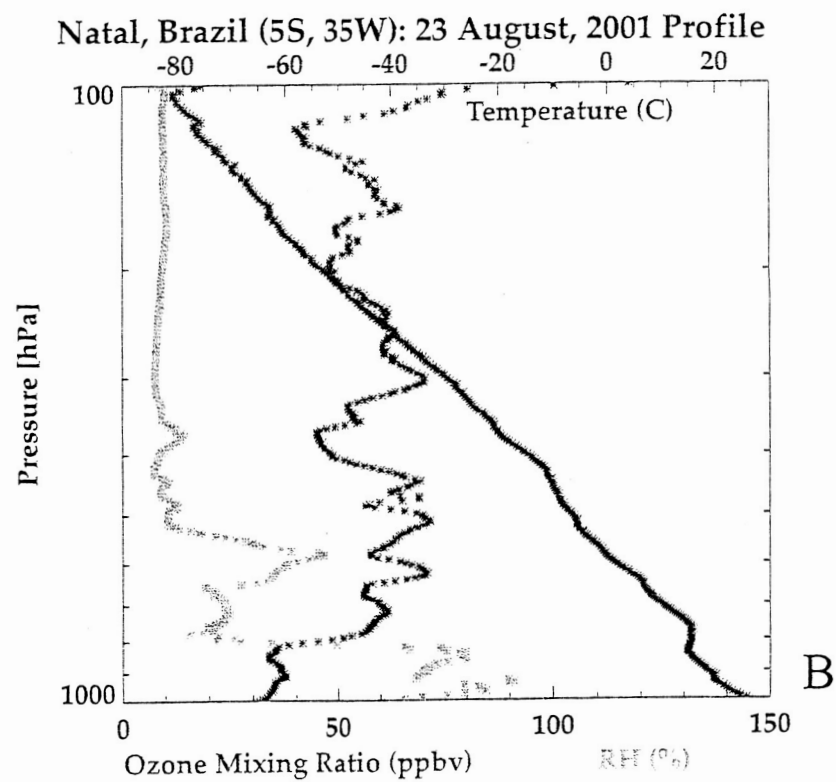
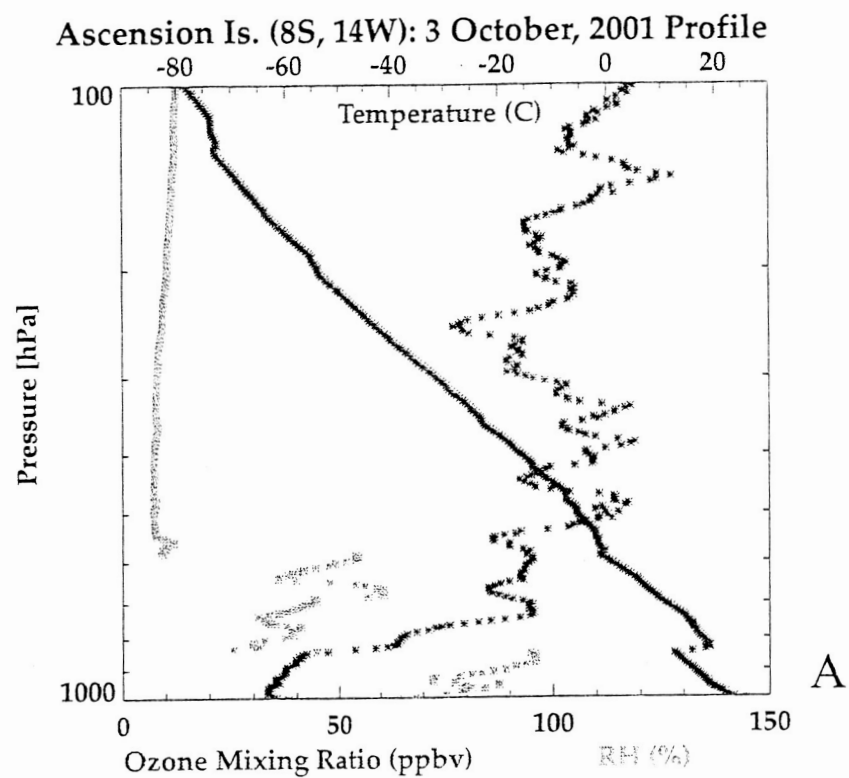
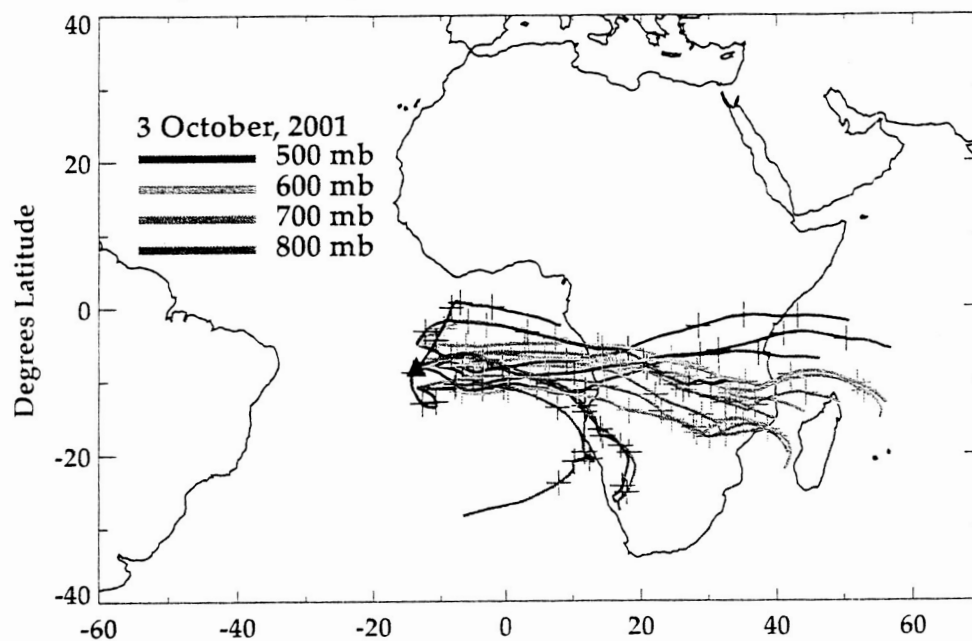


Fig. 7.4-1

8-Day Back-trajectories over Ascension Is. (7S,14W)



8-Day Back-trajectories over Natal (5S,35W)

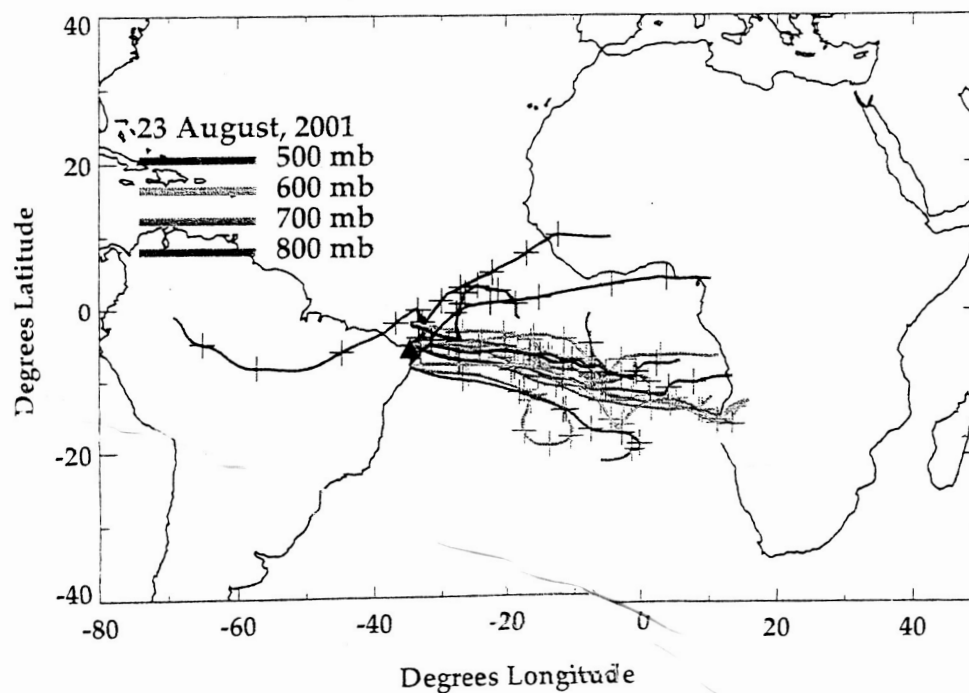
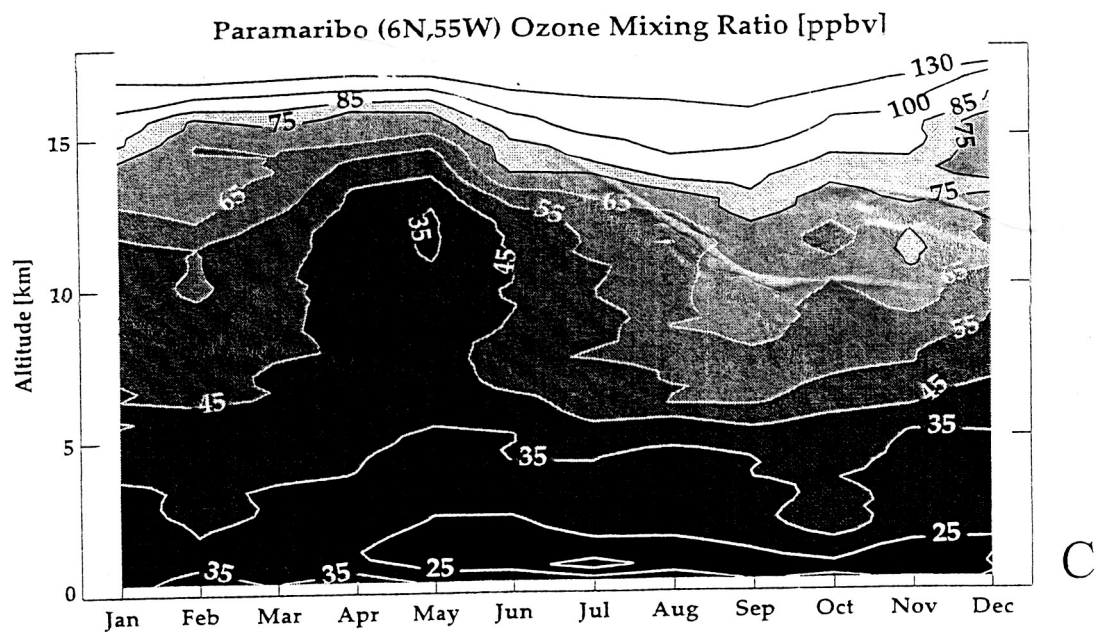
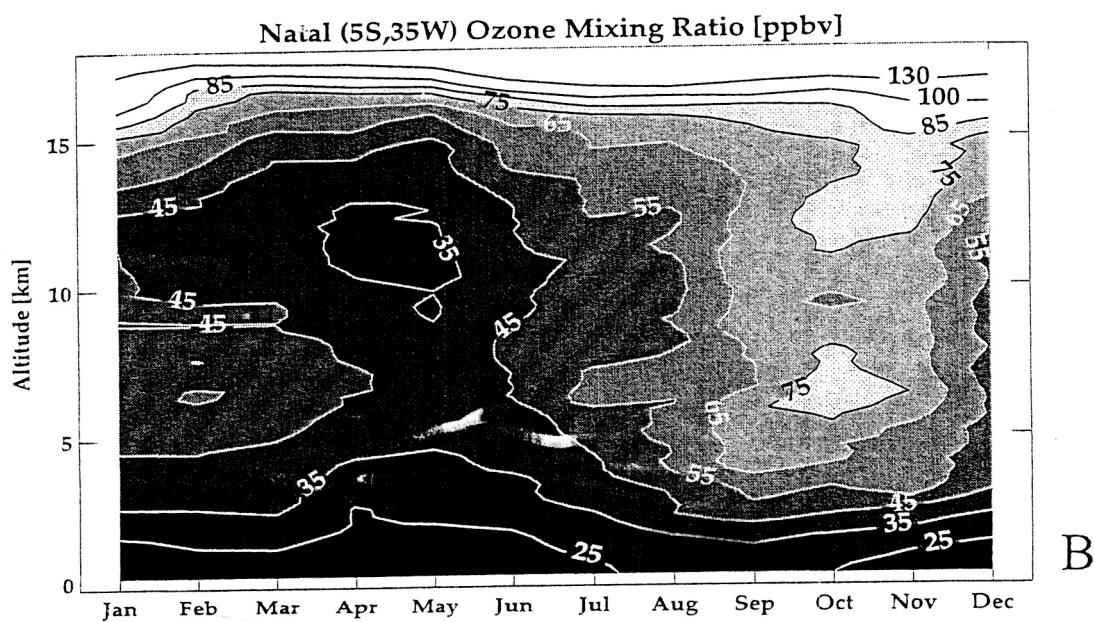
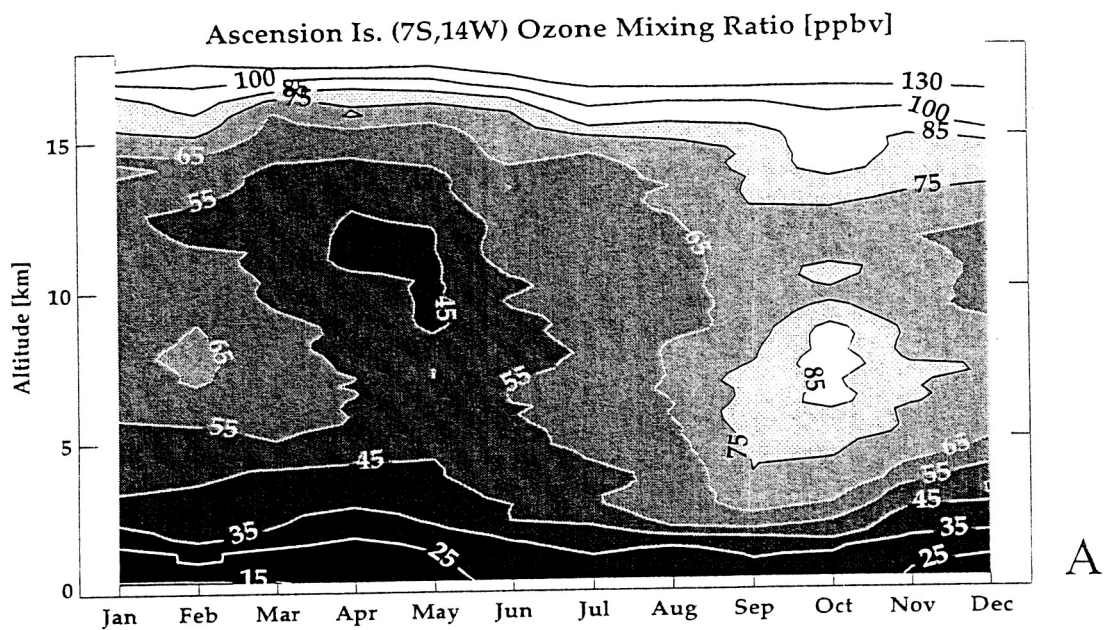


Fig. 7.4-2



Monthly X-sections including years 1998-2002

Fig. 7.4-3

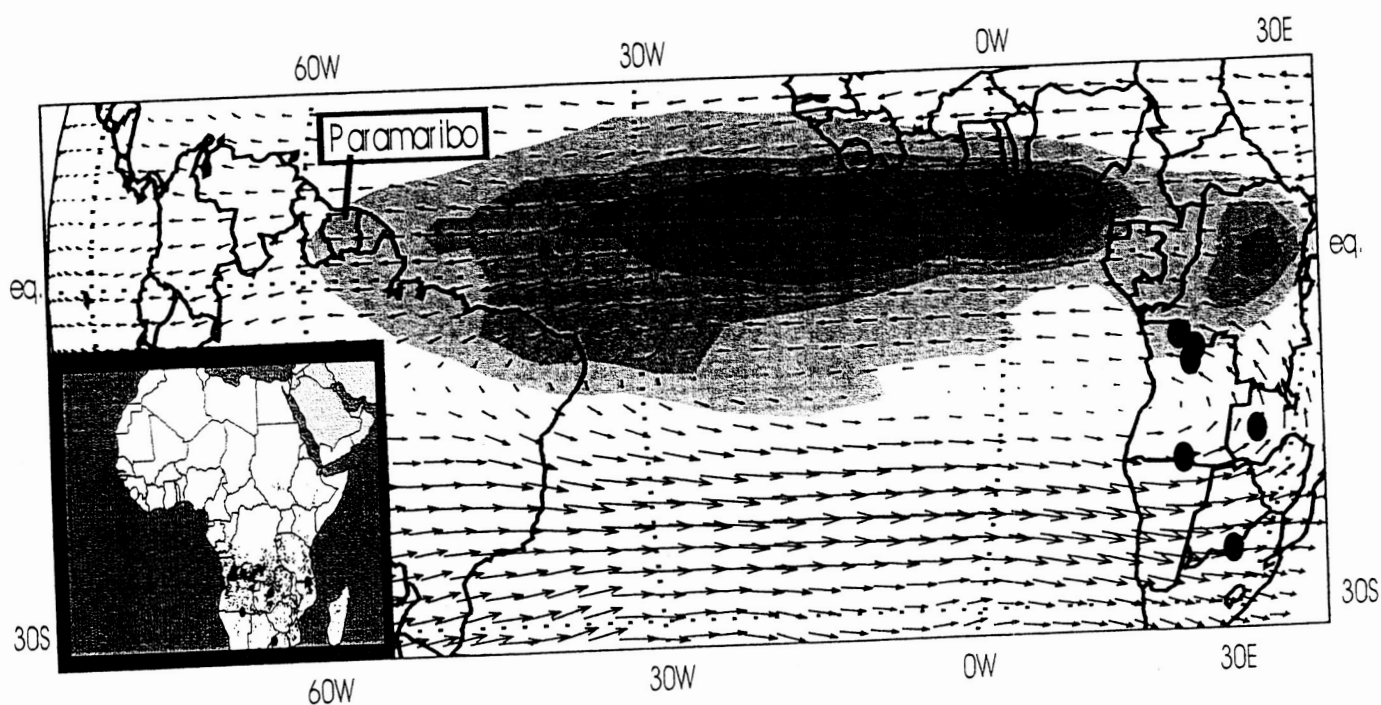


Fig. 7.4-4

Composite illustrating transport of a plume of biomass burning products from Africa. Arrows denote the 10-day average wind at 500hPa from July 31 to August 9, 2000. Filled circles are locations of fires as observed by the ATSR instrument (Arino and Melinotte, 1995, 1998). The insert of Africa shows burning scars from fires observed from METEOSAT (Pinty, 2000). Contours illustrate the dispersion of a modeled tracer released from the fires into the ECMWF wind and transported to Paramaribo by convection and advection. Widespread ozone enhancements were observed in Paramaribo August 9, 2000.

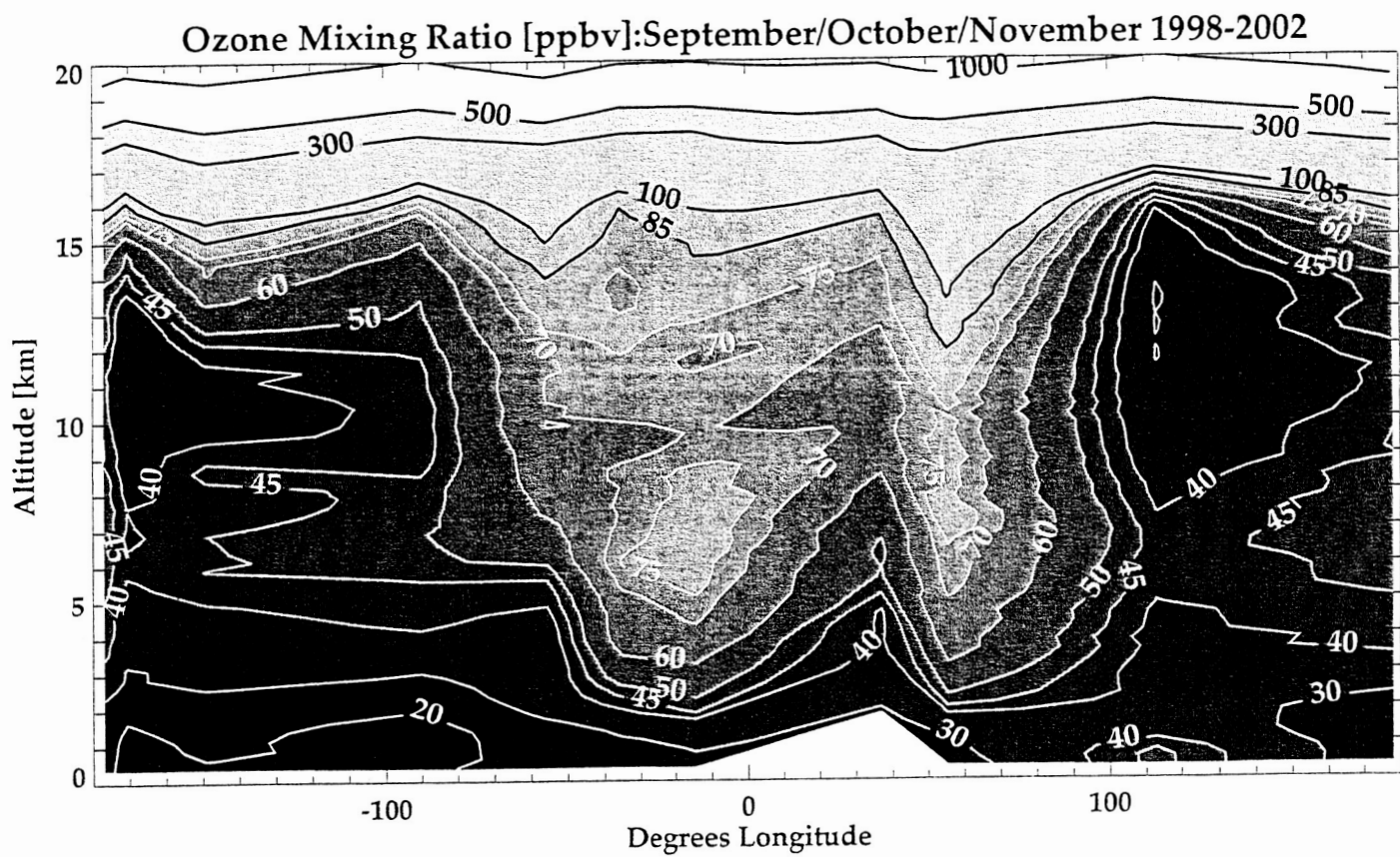


Fig. 7.4-5

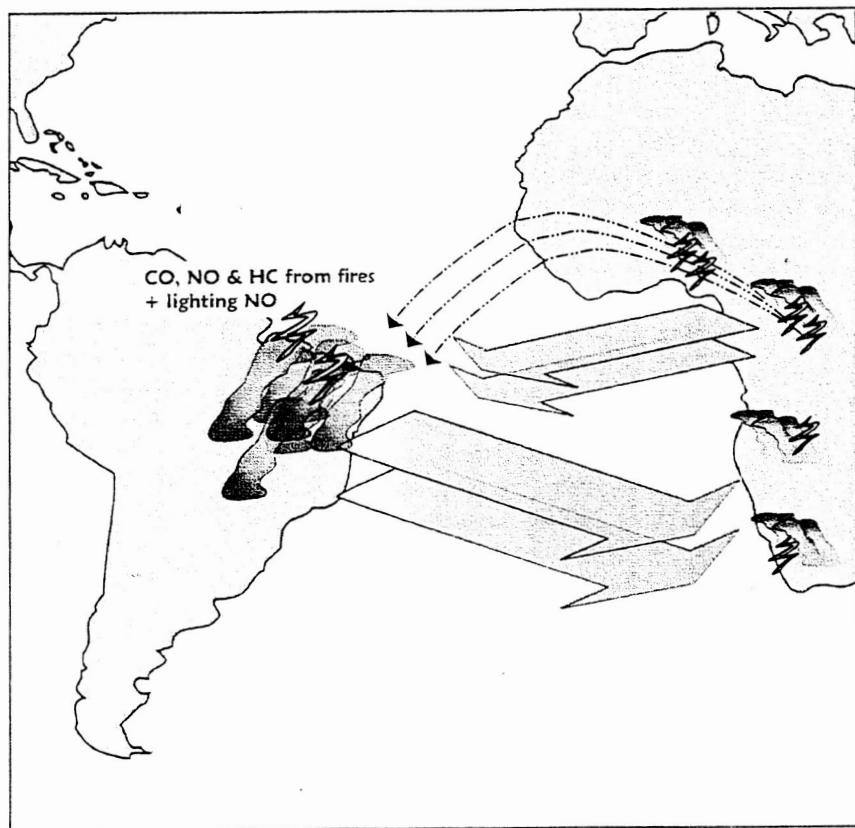


Fig. 7.4-6

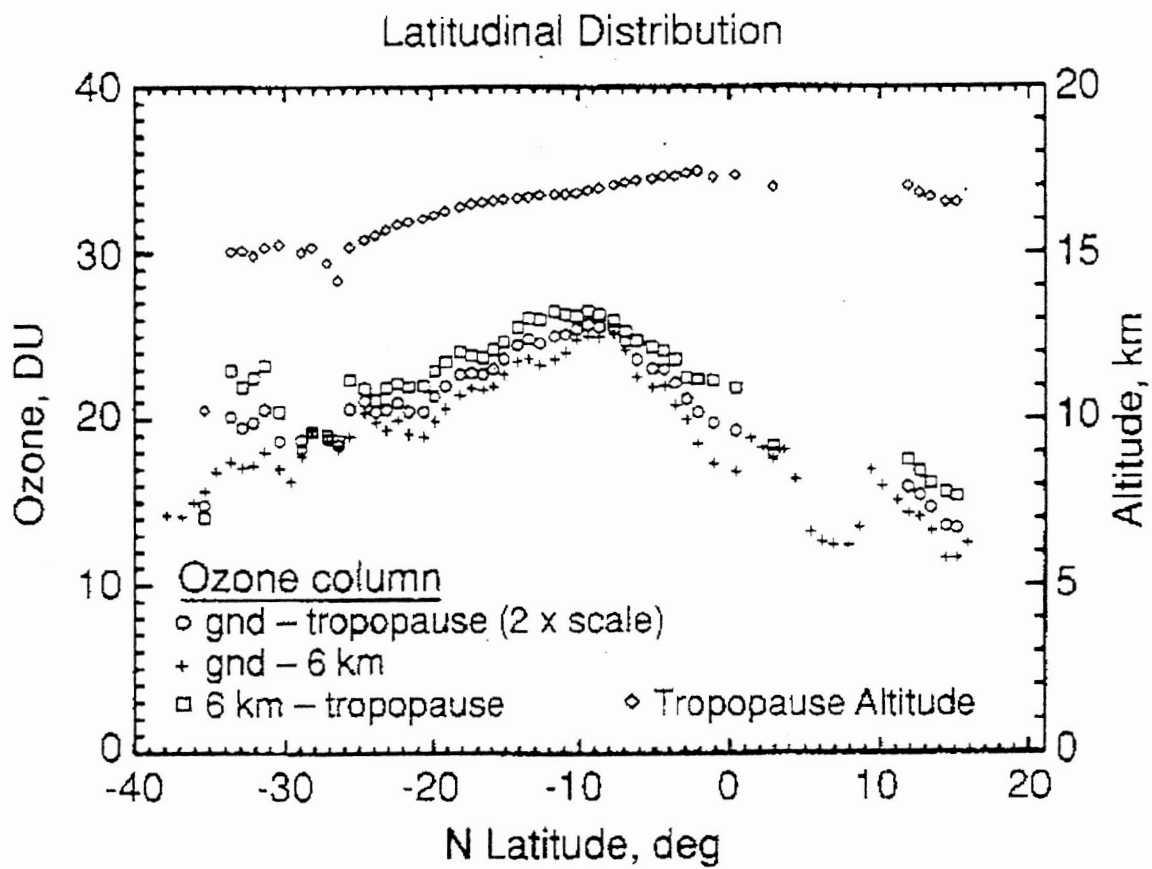


Fig. 7.4-7

MODIFIED RESIDUAL TROPOSPHERIC O3 (DOBSON UNITS)

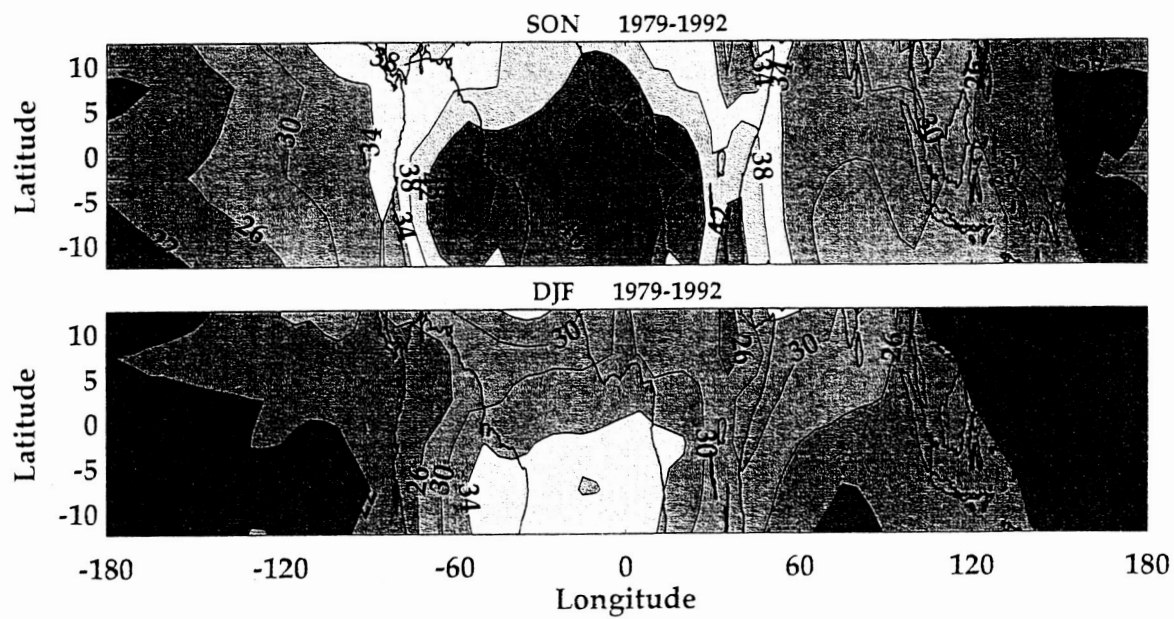


Fig. 7.4-8

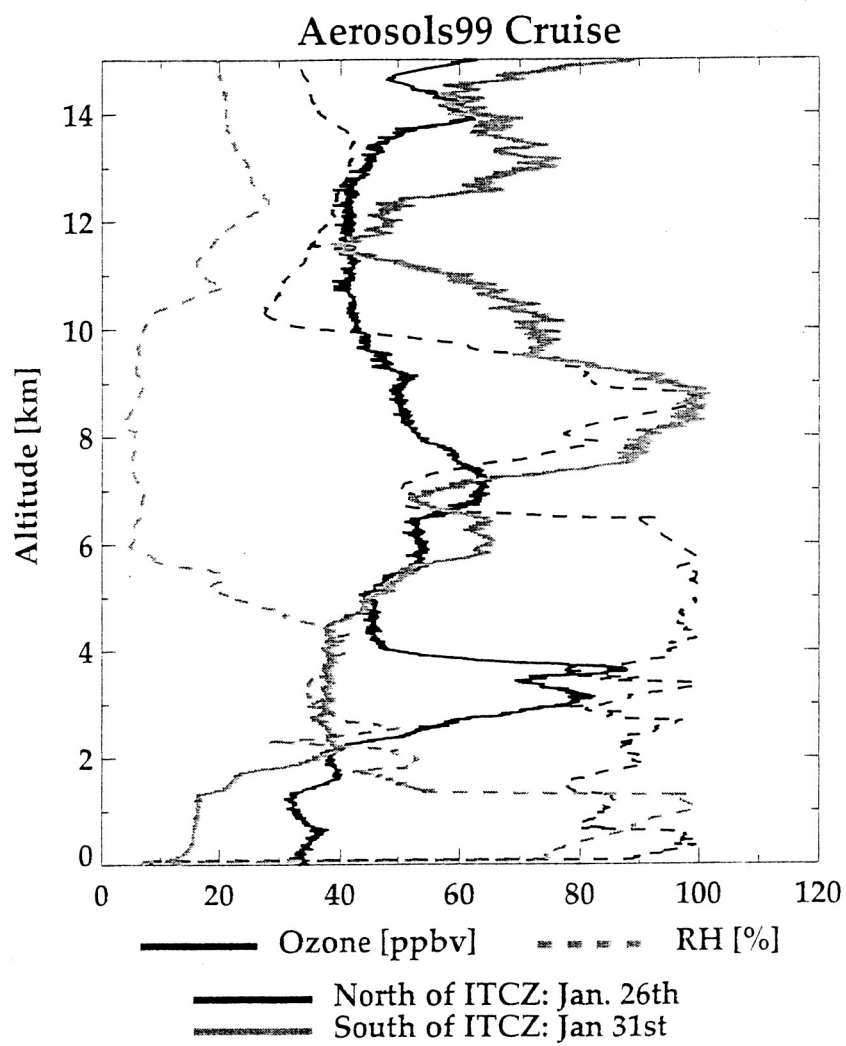


Fig. 7.4-9

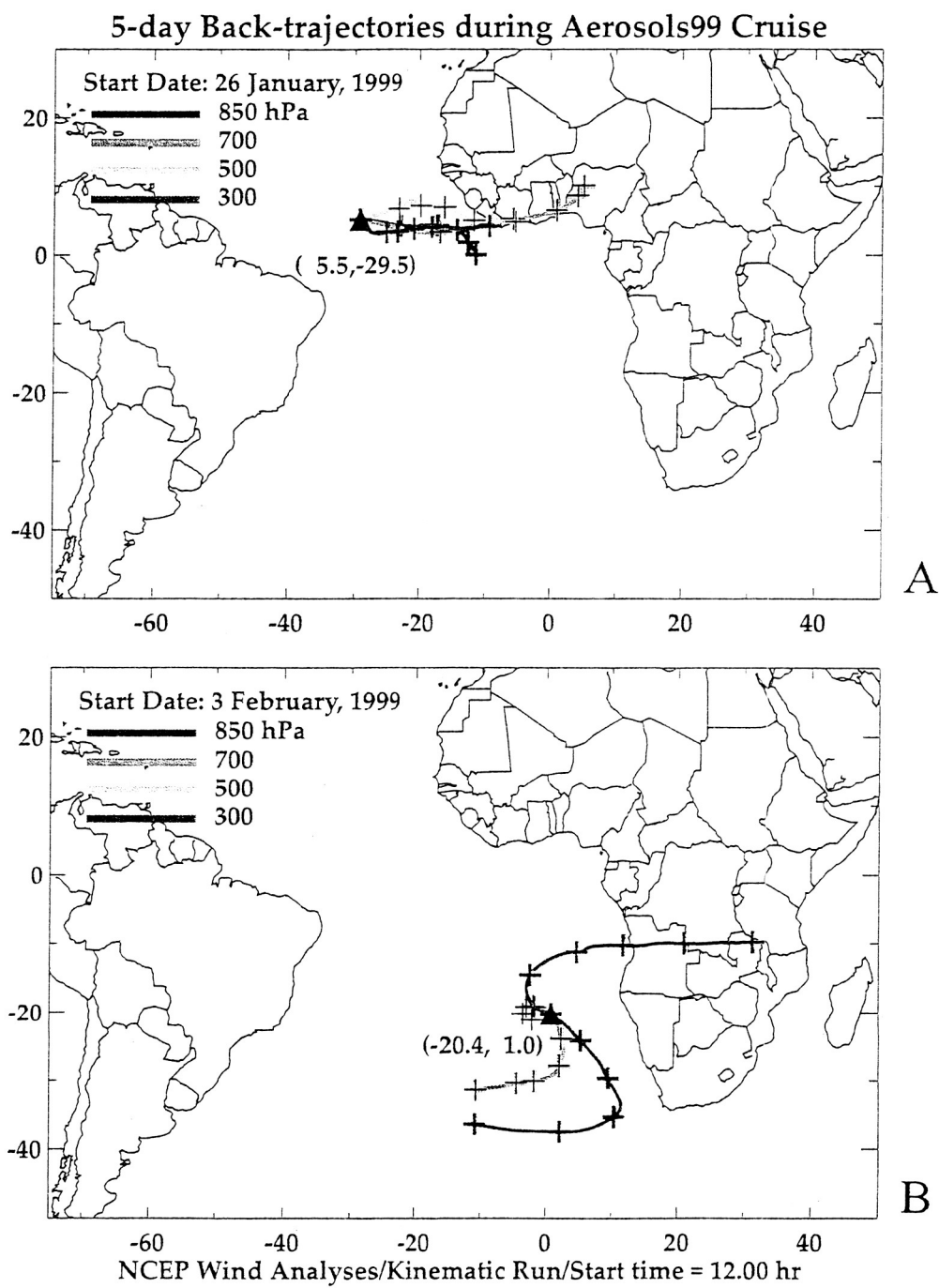
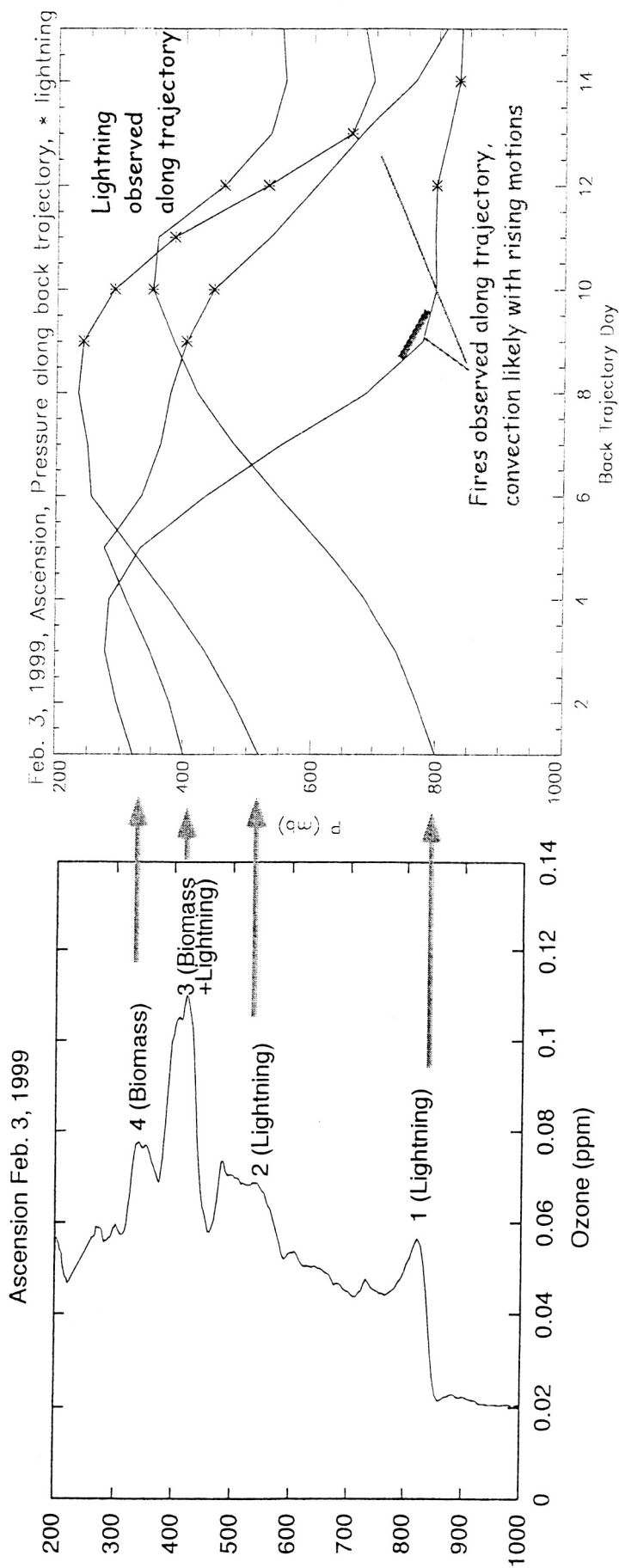


Fig. 7.4-10



African and South American burning produce different patterns of pollution (CO is portrayed here in two cross-sections).

Fig. 7.4-11